



TM5-FASST: a global atmospheric source-receptor model for rapid impact analysis of emission changes on air quality and short-lived climate pollutants

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Abstract This paper describes, documents and validates the TM5-Fast Scenario Screening Tool (TM5-FASST), a global reduced-form air quality source-receptor model that has been designed to compute ambient pollutant concentrations as well as broad range of pollutant-related impacts, related to human health, agricultural crop production, and short-lived pollutant climate metrics, taking as input annual pollutant emission data aggregated at the national or regional level. The TM5-FASST tool, providing a trade-off between accuracy and applicability, is based on linearized emission-concentration sensitivities derived with the full chemistry-transport model TM5. The tool has been extensively applied in various recent critical studies. Although informal and fragmented validation has already been performed in various publications, this paper provides a comprehensive documentation of all components of the model and a validation against the full TM5 model. We find that the simplifications introduced in order to generate immediate results from emission scenarios are not compromising the validity of the output and as such TM5-FASST is proven to be a useful tool in science-policy analysis. Furthermore, it constitutes a suitable architecture for implementing the ensemble of source-receptor relations obtained in the frame of the HTAP modelling exercises, thus creating a link between the scientific community and policy-oriented users.



1 Introduction

There is a need for computationally-efficient methods and tools that provide an integrated environmental assessment of air quality and climate policies, which have a global dimension with sufficient regional detail, and evaluate different impact categories in an internally consistent way. Increasingly, reduced-form source-receptor models are used to compute atmospheric concentrations (and related impacts) from changes in emissions. Source-receptor model studies are available on a regional scale (e.g. GAINS EUROPE, based on the EMEP chemistry-transport model (Amann et al., 2011), GAINS-ASIA, based on the TM5 chemistry-transport model (Amann et al., 2008) and have been proven to be very useful in cost optimization and cost-benefit analysis because of their low computational cost (Amann et al., 2011).

An extensive collaborative global chemistry modelling effort evaluated local and long-range pollutant responses to emission reductions in 4 world regions in the first phase of HTAP (Dentener et al., 2010; Fiore et al., 2009), hereafter referred to as HTAP1. The resulting ensemble source-receptor relations between those regions have been used to evaluate the driving factors behind regional ozone changes in 5 world regions (Wild et al., 2012). Similarly, Yu et al. (2013) evaluated aerosol radiative forcing (RF) from HTAP1 results, while Fry et al. (2012) assessed the RF effects by ozone. Several papers in this special issue (e.g. Stjern et al., 2016) are devoted to advance the HTAP analysis with new models and methodologies.

One of the participating global models in the HTAP1 assessment was the 2-way nested global chemical transport model TM5, applied with $1^\circ \times 1^\circ$ resolution over the continents (Krol et al., 2005). In order to address the need for swift scenario analysis, we used TM5 to develop a reduced-form global source-receptor (SR) model which addresses the issues above, opting for a trade-off between accuracy and speed. The reduced-form version was named “TM5-Fast Scenario Screening Tool” (TM5-FASST). The TM5-FASST approach refines and extends the one developed in the HTAP1 assessment by defining source-receptor regions at a finer resolution and by implementing a direct emission-based calculation of pollutant concentrations and their impacts. To our knowledge such a comprehensive global source-receptor model for a variety of components and impacts (primary and secondary particulate matter, trace gases, wet and dry deposition, climate and health metrics) is at this moment not available for fast impact assessments. The need for models like TM5-FASST is demonstrated by its extensive application in various critical studies (OECD, 2016; Rao et al., 2016; The World Bank, The International Cryosphere Climate Initiative, 2013; UNEP, 2011). An overview of earlier studies with TM5-FASST, in which fragmented and informal validation has already been performed, is given in section S1 of the Supplemental Information (SI).

The tool is undergoing continuous developments and updates regarding metrics and impact evaluations. Hereafter we will refer to the native chemical transport model and the derived SR model as TM5 and TM5-FASST_v0 (or its shortcuts TM5-FASST and FASST) respectively, with version number v0 referring to the features and methodologies described in this paper and as applied in the earlier assessments.

The present paper is a comprehensive documentation of the model and its validation against TM5, to ensure credibility and future applications. In section 2, we describe the methods implemented in TM5-FASST to evaluate in a single framework a broad portfolio of short-lived air pollutants (including CH_4) and their environmental impacts, such as interaction with climate, impact on human health, on natural vegetation and crops. Section 3 focuses on how the derived reduced-form TM5-



FASST replicates the full native TM5 model in terms of linearity, additivity and application to a realistic set of future scenarios. We also evaluate the performance of TM5-FASST against some case studies from literature. We finish with discussing the limitations of the methodology, future development paths and possible ways forward for the best-use of such modelling systems for future policy assessments. Indeed, like the development of TM5-FASST was building on and extending the HTAP1 experiments in a single model context, the regional definitions and sector definitions used in HTAP2 (Galmarini et al., 2017; Koffi et al., 2016) were largely synchronized with the TM5-FASST set-up, increasing the community's capacity for multi-model assessments of hemispheric pollution. It is intended that the lessons-learned are informing the HTAP2 exercise.

2 Methods

2.1 The native TM5 model.

The Tracer Model version 5 (TM5) is a 3-dimensional global atmospheric chemical transport model that simulates the transport, chemical processes, as well as wet and dry deposition of chemically active atmospheric trace gases (e.g. ozone O_3 , SO_2 , NO_x , VOCs, NH_3), and particulate matter components, including SO_4^{2-} , NO_3^- , NH_4^+ , primary $PM_{2.5}$ and its components black carbon, organic carbon, sea salt, and mineral dust. TM5 model version TM5-JRC-cy2-ipcc was used to compute the source receptor relationships as first described by Krol et al., (2005). This model version was used in the PhotoComp scenario studies (e.g. Dentener et al., 2006b; Stevenson et al., 2006) and in the HTAP1 multi-model source receptor assessment (e.g. Anenberg et al., 2009; Fiore et al., 2009; Wild et al., 2012). TM5-JRC-cy2-ipcc (abbreviated TM5) results used in the present study allow comparison with a range of other global model results in HTAP1, but ignore subsequent updates and improvements in TM5 as for instance described in Huijnen et al. (2010), which we consider not critical for this study.

The TM5 model operates with offline meteorology from the European Centre for Medium range Weather Forecasts (ECMWF; 6 hours IFS forecast). These data are stored at a 6-hourly horizontal resolution of $1^\circ \times 1^\circ$ for large-scale 3D fields, and 3-hourly resolution for parameters describing exchange processes at the surface. Of the 60 vertical layers in the operational (OD) ECMWF model (status ca. 2008), a subset of 25 layers is used within TM5, including 5 in the boundary layer, 10 in the free troposphere, and 10 stratospheric layers. Although for most health and ecosystem impacts only the surface level fields are required, base simulation and perturbed pollutants concentrations were calculated and stored for the 25 vertical levels of the model as monthly means, and some air quality-relevant parameters as hourly or daily fields. The meteorological fields are from the ECMWF operational forecast representative for the year 2001.

TM5 utilizes a so-called two-way nested approach, which introduces refinements in both space and time in predefined regions. The nesting comprises a regional high resolution 'zoom' ($1^\circ \times 1^\circ$) within relatively coarse global resolution ($6^\circ \times 4^\circ$), and a transitional grid of $3^\circ \times 2^\circ$. A pre-processing software aggregated the 3D $1^\circ \times 1^\circ$ meteorological fields into the



abovementioned coarser resolutions in a fully mass-conserving way. TM5 has a flexible choice of regional extent and amount of zoom regions. For instance, the HTAP1 simulation setup utilized a set of 4 simultaneous $1^\circ \times 1^\circ$ zooms nested over Europe, North America, South and East Asia. Since hundreds of simulations are needed to drive the TM5-FASST Source-Receptor model, due to computational constraints, it was decided to use single zoom regions, covering the countries and regions for which emission perturbation studies were carried out. For example, the European zoom would contain all European countries, the East Asian zoom region countries like China and Korea, etc. An overview of zoom regions and their regional extent is given in SI section S2. Post-processing software merged the outputs of base and sensitivity simulations into uniform $1^\circ \times 1^\circ$ fields. Some numerical inconsistencies of this merging procedure did occur, but were evaluated to be generally small and occurring over ocean regions, or regions with low population density. We note that at the time of development of the ‘zoom’ model, this specific model set-up allowed to perform photochemistry and aerosol calculations with a relatively high $1^\circ \times 1^\circ$ resolution in the source regions, while other global models were operating at much coarser resolutions of typically T42 ($2.8^\circ \times 2.8^\circ$). With the introduction of massive parallel computing, however, this comparative advantage is now slowly disappearing, and global model resolution of $1^\circ \times 1^\circ$ or finer are now becoming more common (see model descriptions in this special issue). More details on TM5, together an overview of earlier validation efforts is provided in Section S2 of the SI.

2.2 Base emissions

As base simulation emissions we use the community generated representative concentration pathways (RCP) pollutant emissions for the year 2000 at $1^\circ \times 1^\circ$ resolution, prepared for IPCC 5th Assessment (Lamarque et al., 2010). Relevant emitted anthropogenic pollutants include SO_2 , NO_x , NH_3 , black carbon (BC), organic carbon (OC), NMVOC, CO and CH_4 . (Semi-) natural emissions (sea-salt, mineral dust, volcanoes, lightning, vegetation, biomass burning, and terrestrial and oceanic DMS) for the base simulations were included following the recommendations for the AEROCOM study (Dentener et al., 2006a) but they are not affected in the perturbation simulations where we consider only perturbations of anthropogenic.

2.3 Air pollutants source-receptor relations

In general, air quality source-receptor models (AQ-SRM) link emissions of pollutants in a given source region with downwind concentrations and related impacts, implicitly including the underlying effects of meteorology and atmospheric chemical and physical processes. The source region is any point or area from which emissions are considered; the receptor is any point or area at which the pollutant concentration and impact is to be evaluated. Primary pollutants do not undergo chemical transformation during their atmospheric lifetime and are only affected by dry and wet removal from the atmosphere (e.g. elemental carbon, seasalt and mineral dust). Secondary pollutants are formed from reactions of primary emissions, e.g. NO_2 forms nitrate aerosol but also leads to the formation of O_3 ; emitted SO_2 is transformed into sulfate aerosols.

A change of pollutant emissions has the potential to change the chemical formation of other secondary species, e.g. NO_2 affects the oxidative capacity of the atmosphere and therefore influences the lifetime of methane. In summary, a specific secondary component and related impact can be influenced from one or more emitted precursors, and an emitted precursor



can change the impact from one or more parameters. An AQ-SRM will need to include a functional relationship between each precursor and each relevant pollutant or pollutant metric, for each source region and each receptor region.

TM5-FASST_v0 has been designed as a reduced-form SRM: the relation between the emissions of compound i from source x and resulting concentration (or burden) of pollutant j (where $j = i$ in case of a primary component) at receptor y is expressed by a simple functional relation that mimics the underlying meteorological and chemical processes. In the current version v0 of TM5-FASST the function is a linear relation expressing the change in pollutant concentration in the receptor region upon a change in precursor emissions in the source region with the generic form $dC_y = SRC \times dE_x$ where dC_y equals the change in the pollutant concentration compared to a reference concentration in receptor region y , dE_x is the change in precursor emission compared to a reference emission in source region x , and SRC the source-receptor coefficient for the specific compound and source-receptor pair. The source-receptor coefficients are implemented as matrices with dimension $[n_x, n_y]$ with n_x and n_y the number of source and receptor regions respectively. A single SR matrix is available for each precursor and for each resulting component from that precursor. Table 1 gives an overview of all precursor – pollutant links that have been included.

For TM5-FASST_v0 we defined 56 source regions, as shown in Fig. 1. A detailed break-down of regions by country is given in Section S2 of the SI. The choice of regions has been made to obtain an optimal match with integrated assessment models such as IMAGE (Eickhout et al., 2004; van Vuuren et al., 2007), MESSAGE (Riahi et al., 2007), GAINS (Höglund-Isaksson and Mechler, 2005) as well as the POLES model (Russ et al., 2007; Van Aardenne et al., 2007). Most European countries are defined as individual source regions, except for the smallest countries, which have been aggregated. In the current version v0, the USA, China and India are treated as a single emission regions each, i.e. without break-down in states or provinces. Although most integrated assessment models cover Africa, South America, Russia and South-East Asia as a single socio-economic entity, it was decided to sub-divide these regions, to account for climatological difference in these vast continents. Apart from the 56 regions, source-receptor coefficients were calculated between global international shipping and aviation as sources, and the global grid as receptor, resulting in $n_x = 58$ source functions.

The SR matrices, describing the concentration response in each receptor upon a change in emissions in each source region, have been derived from a set of simulations with the full chemical transport model TM5 by applying -20% emission perturbations for each of the 56 defined source regions (plus shipping and aviation), for all relevant precursor components, in comparison to a set of unperturbed simulations, hereafter denoted as ‘base simulations’. A perturbation of -20% is consistent with the approach in HTAP1. It is small enough to evaluate emission-concentration sensitivities under present day conditions, while still allowing an extrapolation to larger emission changes in scenarios of the future. As elucidated in previous section base and perturbed simulations are available on a $1^\circ \times 1^\circ$ global resolution. Figures S3.1 and S3.2 in the SI shows some examples of SR grid maps for $PM_{2.5}$, O_3 metrics, deposition and column burden for source regions China, India and USA, illustrating clearly the difference in long-range transport characteristics between different species.



For each receptor point y (i.e. each model vertical level $1^\circ \times 1^\circ$ grid cell), the change in concentration of component i in receptor y resulting from a -20% perturbation of emitted precursor j in source region x , is expressed by a unique SR coefficient $A_{ij}[x, y]$:

$$A_{ij}[x, y] = \frac{\Delta C_j(y)}{\Delta E_i(x)} \text{ with } \Delta E_i(x) = 0.2 E_{i,base}(x) \quad (1)$$

- 5 The total concentration of component j in receptor region y , resulting from arbitrary emissions of *all* n_i precursors i at *all* n_x source regions x , is obtained as a perturbation on the base-simulation concentration, by summing up all the respective SR coefficients scaled with the actual emission perturbation:

$$C_j(y) = C_{j,base}(y) + \sum_{n_x} \sum_{n_i} A_{ij}[x, y] \cdot [E_i(x) - E_{i,base}(x)] \quad (2)$$

- 10 Pollutants C_j include particulate matter components (SO_4 , NO_3 , NH_4 , BC, particulate organic matter – POM), trace gases (SO_2 , NO, NO_2 , NH_3 , O_3), and deposition fluxes of BC, N and S species. In the case of ozone, the n_i precursors in equation (2) would comprise [NO_x , NMVOC, CO, CH_4]. The set of linear equations (2) with associated source-receptor matrices (1) for all components and all source and receptor regions thus emulates the ‘full’ TM5-CTM, and constitutes the ‘kernel’ of TM5-FASST_V0. When OC emissions are provided in mass units C, the OC mass is multiplied with a factor 1.3 to obtain Particulate Organic Matter (POM) (Kanakidou et al., 2005).

- 15 BC and POM emissions are assumed not to interact with other pollutants and their atmospheric lifetime are assumed not to be affected by mixing with other soluble species like sulfate, nitrate or ammonium salts. We note that, unlike many other inventories, the RCP emission scenarios do not include a separate inventory for total primary PM2.5 which includes besides BC and POM other non-specified primary particulates (e.g. primary sulfate, fly-ash). When specific scenario studies require so, TM5-FASST_v0 treats this ‘other’ primary PM.5 (OPP = Primary PM2.5 – BC – POM) as BC in Eq. (2), where both
 20 $C_{OPP,base}$ and $E_{OPP,base}$ are zero.

$$C_{OPP}(y) = \sum_{n_x} \sum_{n_i} A_{BC}[x, y] \cdot E_{OPP}(x)$$

Secondary biogenic POM (SOA) was included following the AEROCOM recommendation (Dentener et al., 2006a; Kanakidou et al., 2005) which parameterized SOA formation from natural VOC emissions as a fixed fraction of the primary emissions. SOA from anthropogenic emission was not explicitly included in the current simulations. This is a topic for future developments of the model.

- 25 In TM5-FASST_v0 the monthly perturbations are aggregated to annual emission-concentration SR matrices, as the health, climate and vegetation impact metrics used in this version are also aggregated to annual values. Surface ozone (and NO_2) fields were stored at hourly intervals allowing for the calculation of specific vegetation and health related O_3 metrics, often based on thresholds of hourly O_3 concentrations, or concentrations during daytime. The hourly O_3 surface fields were converted into specific O_3 metrics responses to annual emissions, including accumulated hourly ozone above a threshold of
 30 40 ppbV during a 3 months crop growing season (AOT40), 3-monthly mean of 7 hr or 12 hr daytime ozone during crop



growing season (M7, M12), maximum 6-monthly running average of daily maximum hourly O_3 (M6M), the sum of daily maximal 8hr ozone mean concentrations above 35ppbV (SOMO₃₅).

The -20% emission perturbation calculations were performed for the combination of emission perturbations given in Table 2. For a limited set of representative source regions, an additional wider range of emission perturbations [-80% to +100%] has been applied to evaluate possible non-linearities in the emission-concentration relationships. The list of these additional perturbation simulations is given in Table S3 of the SI.

We did not perform dedicated perturbation simulations on CH_4 as O_3 precursor, but implemented TM5 results obtained in the frame of the first phase of the Hemispheric Transport of Air Pollutants (HTAP1) assessment (Dentener et al., 2010; Fiore et al., 2008). In one of the prescribed experiment set-up, models evaluated how surface ozone levels are responding when the global steady-state CH_4 concentration decreases with 20% from 1760 ppbv (the global mean CH_4 concentration in the year 2000) to 1408 ppbv. The outcome of this experiment is a set of global grid maps with hourly O_3 concentration responses from which all relevant O_3 metrics can be obtained. As an example, the annual mean O_3 concentration response to the CH_4 perturbation is shown in Fig. S3.3 in the SI. We note that the HTAP1 CH_4 perturbation experiment is not a set-up that evaluates the ozone response to a change in CH_4 emissions which would involve large transient time-scale computations, but rather the steady-state result of an established changed background CH_4 concentration. The change in CH_4 burden due to OH oxidation under this prescribed concentration change is treated as an emission perturbation, in the case of TM5 corresponding to a sustained emission perturbation of 77Tg/year, allowing to normalize the resulting responses in O_3 concentration and metrics on a CH_4 emission basis which are then linearly scaled in the reduced-form TM5-FASST set-up. Because of its long life time compared to short-lived ozone precursors, CH_4 source-receptor coefficients are considered independent on the location of emission and are therefore provided as global emission-to-regional (or gridded) concentration responses.

Because of the mismatch between the HTAP1 source - receptor regions and the FASST ones, the current version of TM5-FASST does not include source-receptor relations between CO and O_3 concentration (or O_3 exposure metrics), only impacts of CO emissions on global methane and O_3 global radiative forcing, also in this case retrieved from HTAP1 dedicated CO perturbation experiments with TM5. Although the impact of CO on O_3 concentration is limited - based on the HTAP1 CO perturbation simulations with TM5, we estimate that anthropogenic CO emissions contribute with 1 – 1.9 ppbv in annual mean O_3 over Europe, 1.3 -19 over North-America, 0.7-1 over South Asia and 0.3 – 1.5 over East-Asia – this is an important issue for the further development of the tool.

Deposition source-receptor matrices of nitrogen and sulfur compounds are obtained in the same way as for the pollutant ambient concentration fields, making the difference between the base and perturbation simulations. Nitrogen depositions are calculated from accumulation of the instantaneous surface budgets of all relevant nitrogen components (NO , NO_2 , NO_3 , $2 \times N_2O_5$, HNO_4 , organic nitrates, NH_3 , NH_4) and similar for sulfur from SO_2 and SO_4 , into monthly time steps. Column amounts of ozone and particulate matter are also computed using 3D monthly output of concentrations and meteorological parameters.



2.4 Sub-grid PM_{2.5} gradients

TM5-FASST is specifically aiming at providing pollutant exposure fields for further impact evaluation. For the evaluation of health impacts from outdoor air pollution, a 1°x1° horizontal resolution may not adequately represent sub-grid gradients of pollutants. In the current study we only consider PM_{2.5}, although also ozone and NO₂ are likely subject to sub-grid gradients. Indeed, higher pollutant levels are expected to concur with high population density in urban areas, hence an area-averaged concentration for a nominally 100x100km² sized grid cell will underestimate the exposure of population located in pollution hotspots within a single grid cell. We provide a simple parameterization, generating a correction factor on the gridbox area-mean PM_{2.5} concentration, to better represent the actual mean population exposure within that grid cell. The parameterization is based on the underlying assumption that the spatial distribution of *primary emitted* PM_{2.5} correlates with population density. Our parameterization builds upon high-resolution population grid maps, allowing a sub-grid readjustment of the PM_{2.5} concentration within each 1°x1° grid cell. Further it needs additional information to flag the population sub-grids as ‘urban’ or ‘rural’, e.g. population density for which an urban threshold can be defined, or more sophisticated schemes defining urban areas. We further assume that only primary PM_{2.5} from the residential and the surface transport sectors is contributing to the local (urban) increment, while other aerosol precursor components and other sectors are assumed to be homogeneously distributed over the 1°x1° grid cell. Indeed, secondary PM_{2.5} is formed over longer time scales and therefore deemed to be more homogeneously distributed at the regional scale. The adjusted population-weighted mean concentration within each 1°x1° grid cell (conserving the area-based grid cell mean) is then calculated as follows:

$$\text{PM}_{2.5,\text{inc}} = \text{DU} + \text{SS} + \text{SO}_4^{2-} + \text{NO}_3^- + \text{NH}_4^+ + (1-k_{\text{BC}}) \text{BC} + (1-k_{\text{POM}}) \text{POM} + \text{INCR}(k_{\text{BC}} \text{BC} + k_{\text{POM}} \text{POM})$$

with DU and SS the fixed natural mineral dust and sea-salt contributions respectively; SO₄²⁻, NO₃⁻, NH₄⁺, BC and POM the 1°x1° grid cell average values resulting from TM5 or TM5-FASST; k_{BC} (k_{POM}) the fraction of (residential + transport) BC (POM) emissions in the total BC (POM) emissions within the 1°x1° grid cell and INCR the urban increment factor. This sub-grid parameterization has been applied as a part of the methodology to estimate population exposure in the Global Burden of Disease assessments (Brauer et al., 2012). Supplemental Information section S4 provides details on the calculation of INCR.

The required gridded sectorial emission data may not be readily available for any assessment. A “default” set of regional population-weighted averaged increment factors for BC and POM is given in Table S4.2, based on the baseline simulations performed with TM5 for the year 2000, i.e. using year 2000 population (CIESIN GWPv3).

2.5 Health impacts

TM5-FASST provides output of annual mean PM_{2.5} and O₃ health metrics (3-monthly and 6-monthly mean of daily maximum hourly O₃ (M3M, M6M), and the sum of the maximal 8-hourly mean above a threshold of 35 ppbV (SOMO35) or without threshold (SOMO0), as well as annual mean NO_x and SO₂ concentrations at grid resolution of 1°x1°. These are the metrics consistent with underlying epidemiological studies (Jerrett et al., 2009; Krewski et al., 2009; Pope et al., 2002). The



population-weighted pollutant exposure metrics grid maps, in combination with any consistent population grid map, are thus available for human health impact assessment. The TM5-FASST_v0 tool provides a set of standard methodologies, including default population and health statistics, to quantify the number of air quality-related premature deaths from PM_{2.5} and O₃.

5 Health impacts from PM_{2.5} are calculated as the number of annual premature mortalities from 5 causes of death: ischemic heart disease (IHD), chronic obstructive pulmonary disease (COPD), stroke, lung cancer (LC) and acute lower respiratory airways infections (ALRI) whereas mortalities from exposure to O₃ are related to respiratory disease.

Cause-specific excess mortalities are calculated at grid cell level using a population-attributable fraction approach as described in Murray et al. (2003) from $\Delta Mort = m_0 \times AF \times Pop$, where m_0 is the baseline mortality rate for the exposed population, $AF = 1 - 1/RR$ is the fraction of total mortalities attributed to the risk factor (exposure to air pollution), RR =
 10 relative risk of death attributable to a change in population-weighted mean pollutant concentration, and Pop is the exposed population (adults ≥ 30 years old, except for ALRI for which infant population < 5 years old was considered). Sources for currently implemented population and baseline mortality statistics and their projections in TM5-FASST_v0 are given in section S5 of the SI.

15 TM5-FASST currently includes two approaches from the literature to evaluate RRs for PM_{2.5}: the first one follows methodology and outcomes of the American Cancer Society (ACS) study (Krewski et al., 2009; Pope et al., 2002) based on a log-linear exposure response function $RR = \exp^{\beta \Delta PM_{2.5}}$ where β is the concentration–response factor (CRF; i.e., the estimated slope of the log-linear relation between concentration and mortality) and $\Delta PM_{2.5}$ is the change in concentration. The fraction of the disease burden attributable to PM_{2.5} as a risk factor, the attributable fraction (AF), is defined as

$$AF = \frac{RR - 1}{RR} = 1 - \exp(-\beta \Delta PM_{2.5})$$

20 A 10 $\mu\text{g}/\text{m}^3$ increase in PM_{2.5} (concentration range, 5.8–22.2 $\mu\text{g}/\text{m}^3$) was associated with 13% (95% CI, 10–16%), and 14% (95% CI, 6–23%) increases in cardiopulmonary (CP) and lung cancer (LC) mortality, corresponding to β (for a 1 $\mu\text{g}/\text{m}^3$ increase in PM_{2.5}) of 0.01213 and 0.01284 for CP and LC respectively. We also include an evaluation based on total non-accidental mortality with a RR per 10 $\mu\text{g}/\text{m}^3$ of 6.2% (95% CI, 4.0–8.3%) (Hoek et al., 2013).

The second methodology uses age-averaged Integrated Exposure-Response functions (IER) developed by Burnett et al.
 25 (2014), and first applied in e.g. the Global Burden of Disease study (Lim et al., 2012). IERs expand epidemiological studies on the long-term effects of ambient PM_{2.5} exposure to higher concentration ranges than the one available from the ACS study, making use of health impact studies for smoking and second-hand smoking. This tends to flatten off the RR function at high PM_{2.5} concentration levels compared to the traditionally-used log-lin function which, extrapolated outside the concentration range where the health impacts were determined, would lead to unrealistically high mortality fractions
 30 attributed to air pollution. The RR functions are given by:



$$RR(PM_{2.5}) = 1 + \alpha \left[1 - e^{-\gamma(PM_{2.5} - zcf)^\delta} \right] \quad \text{for } PM_{2.5} > zcf$$

$$RR = 1 \quad \text{for } PM_{2.5} \leq zcf$$

where zcf is the counterfactual concentration (theoretical minimum-risk exposure, assumed by Burnett et al. (2014) to have a uniform distribution between 5.8 and 8.8. We used the age-averaged values for parameters α , γ , δ and zcf reported by Burnett et al. (2014) for 1000 simulations (IHME, 2011) to generate a look-up table of cause-specific RRs as a function of $PM_{2.5}$, where for each $PM_{2.5}$ value the mean (of 1000) RRs was used as central value, and the 95% CI as uncertainty range.

Alternatively, we fitted a set of α , γ , δ and zcf parameters to the IER functional shapes based on the generated ($PM_{2.5}$, RR) look-up table. More details on the fitting procedure are given in section S5.3 of the SI, giving also the obtained fitting parameters for aged-average cause-specific RRs.

We note that WHO (2013) recommends both a log-linear and IER approach for long-term mortality from $PM_{2.5}$ exposure, with the log-linear RR referring to total (non-accidental) mortalities rather than the 2 specific causes (CP and LC) in the Krewski et al. (2009) approach. The WHO recommendations however refer specifically to European impact assessments. We deem that the attribution of mortalities to air pollution, as a fraction of total mortalities, rather than attributed to specific diseases, induces large uncertainties in other world regions because of different relative contributions of pollution-related diseases to total mortality

For O_3 exposure, $RR = e^{\beta(\Delta M6M)}$, β is the concentration–response factor, and $RR = 1.040$ [95% confidence interval (CI): 1.013, 1.067] for a 10 ppb increase in M6M according to Jerrett et al. (2009). We apply a default counterfactual concentration of 33.3 ppbV, the minimum M6M exposure level in the Jerrett et al. (2009) epidemiological study.

The coefficients in the IER functions used in the GBD assessments have been recently updated due to methodological improvements in the curve fitting, leading to generally higher RR and mortality estimates (Cohen et al., 2017; Forouzanfar et al., 2016). In particular, the theoretical minimum risk exposure level was assigned a uniform distribution of 2.4–5.9 $\mu\text{g}/\text{m}^3$ for $PM_{2.5}$, bounded by the minimum and fifth percentiles of exposure distributions from outdoor air pollution cohort studies. Further, a recent health impact assessment (Malley et al., 2017), using updated RR estimate and exposure parameters from the epidemiological study by Turner et al. (2016), estimates 1.04–1.23 million respiratory deaths in adults attributable to O_3 exposure, compared with 0.40–0.55 million respiratory deaths attributable to O_3 exposure based on the earlier (Jerrett et al., 2009) risk estimate and parameters. These updates have not been included in the current version of TM5-FASST.

The inclusion of a theoretical minimum risk exposure level (zcf) in the $PM_{2.5}$ exposure-response functions is motivated by the lowest prevailing concentration at which an increased risk was observed in the ACS cohort studies. Burnett et al. (2014) argue that zero exposure is not a practical counterfactual level because it is impossible to achieve even in pristine environments, implicitly indicating that their exposure-response curves strictly apply to total $PM_{2.5}$, including the natural components (mineral dust, sea-salt). In impact assessment studies, evaluating the difference between two anthropogenic emission scenarios (under otherwise identical natural background conditions) is often more relevant than evaluating absolute impacts for a single scenario. Therefore, TM5-FASST includes the option to customize the value of zcf , both in the IER as



the log-linear shaped functions. In practice, we recommend to use $zcf=0$ when evaluating anthropogenic emissions only. Because of the non-linear IER functions, Δ mortalities between 2 scenarios (S1, S2) with population-weighted $PM_{2.5}$ concentrations PM_{S1} and PM_{S2} respectively are evaluated as $Mort(PM_{S2}) - Mort(PM_{S1})$, and not as $Mort(PM_{S2}-PM_{S1})$. Health impacts from exposure to other pollutants (NO_2 , SO_2 for example) are currently not being evaluated in TM5-FASST-v0 although the model output does provide population-weighted mean concentrations of NO_x and SO_2 .

2.6 Crop impacts

The methodology applied in TM5-FASST to calculate the impacts on four crop types (wheat, maize, rice, and soy bean) is based on Van Dingenen et al. (2009). In brief, TM5 base and -20% perturbation simulations of gridded crop O_3 exposure metrics (averaged or accumulated over the crop growing season) are overlaid with crop suitability grid maps to evaluate receptor region-averaged exposure metrics SR coefficients.

Available metrics are accumulated ozone above 40 ppbV (AOT40) and seasonal mean 7 hr or 12 hr day-time ozone concentration (M7, M12) for which exposure-response functions are available from the literature (Mills et al., 2007; Wang and Mauzerall, 2004).

Both M_i metrics are calculated as the 3-monthly mean daytime (09:00 – 15:59 for M7, 08:00 – 19:59 for M12) ozone concentration, evaluated over the 3 months centred on the midpoint of the location-dependent crop-growing season. The Weibull-type exposure-response functions express the crop relative yield (RYL) loss as a function of M_i :

$$RYL = 1 - \frac{\exp\left[-\left(\frac{M_i}{a}\right)^b\right]}{\exp\left[-\left(\frac{c}{a}\right)^b\right]}$$

The parameter values in the exposure response functions and the applied methodology are described in detail by Van Dingenen et al. (2009), however gridded crop data (growing season and suitability, based on average climate 1961 – 1990) have been updated using Global Agro-Ecological Zones data set (IIASA and FAO, 2012, available at <http://www.gaez.iiasa.ac.at/>). Again we note that the non-linear shape of the $RYL(M_i)$ function requires the ΔRYL for 2 scenarios (S1, S2) being evaluated as $RYL(M_{i,S2}) - RYL(M_{i,S1})$, and not as $RYL(M_{i,S2} - M_{i,S1})$.

Finally, it is important to note that TM5-FASST modelled O_3 surface concentrations refer to the middle of the TM5's lower layer gridbox, i.e. 30m above surface, whereas monitoring of O_3 (from which exposure metrics are derived) actually happens at a standard altitude of 3 to 5m above the surface where, due to deposition and meteorological processes, the concentration may differ. However comparing TM5 simulated gridbox-centre ozone metrics with observations from 99 monitoring stations worldwide, Van Dingenen et al. (2009) find that, averaged over the horizontal resolution of the grid cells, the TM5 simulated 30m monthly O_3 and O_3 metrics represent the observed values within their variability range.

2.7 Climate metrics

We make use of the available 3D aerosol fields in the -20% emission perturbation simulations with TM5 to derive the change in global forcing for each of the perturbed emitted precursors. For each emitted pollutant (primary and secondary) the



resulting normalized global forcing responses [W/m^2]/[kg/yr] are then further used to calculate the global warming potential (GWP) and global temperature potential (GTP) for a series of time horizons H . In this way, a set of climate metrics is calculated with a consistent methodology as the air quality metrics, health and ecosystem impacts calculated from the concentration and deposition fields. In this section we describe in more detail the applied methodologies.

5 2.7.1 Instantaneous radiative forcing by aerosols

The base simulation and -20% perturbation response of the column-integrated aerosol mass over all 25 vertical layers of TM5 for all relevant species was calculated and stored. The calculation of the top-of-atmosphere (TOA) instantaneous forcing by aerosol is based on the radiative transfer model described by Marmer et al. (2007) using monthly average meteorological fields and surface characteristics using ECMWF monthly average meteorological fields (temperature, clouds, relative humidity, surface albedo) for the year 2001. Enhanced aerosol scattering of solar radiation back into space increases the planetary albedo and is therefore associated with cooling. On the other hand BC is a strong absorber of solar radiation and is therefore associated with warming (Hess et al., 1998). We assume externally mixed aerosols and calculate the forcing separately for each component (see section S6 of the SI for more details on the forcing calculations). The total aerosol forcing is obtained by summing up these contributions. To avoid further extensive radiative transfer calculation, monthly-mean radiative forcing efficiencies, expressed as [W/m^2]/[μg], were calculated once using the $1^\circ \times 1^\circ$ gridded TM5 base simulation outputs and off-line radiative code using monthly fields of aerosol, ECMWF meteorology and surface characteristics, and stored for further use (Marmer et al., 2007). The annual TOA global forcing for each scenario is then obtained by multiplying the monthly column-integrated aerosol mass with this grid-cell specific monthly mass forcing efficiency and subsequently averaged over one year. Although this method has some limitations, as discussed in Stjern et al. (2016), we demonstrate in the validation section that in the context of the reduced-form FASST approach, the applied method provides reliable results. Figure S6.1 (a,b,c) in the SI shows the resulting global radiative forcing fields for sulfate, POM and BC.

2.7.2 Indirect aerosol forcing

Aerosols modify the microphysical and radiative properties and lifetime of clouds, commonly denoted as the aerosol indirect effect (Haywood and Boucher, 2000). This forcing results from the ability of the hygroscopic particles to act as (warm) cloud condensation nuclei thus altering the size, the number and the optical properties of cloud droplets (Twomey, 1974). More and smaller cloud droplets increase the cloud albedo, which leads to cooling. In this study we have only considered the so far best studied first indirect effect using the method described by Boucher and Lohmann (1995). In particular the parameterization uses the cloud information (liquid water content and cloud cover) from the driving ECMWF ERA data. The cloud droplet number concentrations were calculated the set of equations ‘A’ in Boucher and Lohmann (1995) separating continental and maritime clouds. The cloud droplet effective radius is calculated from the mean volume cloud droplet radius



using equation 4 in Boucher and Lohmann (1995). The global indirect forcing field associated with sulfate aerosols is shown in Fig. S6.1(d) of the SI.

2.7.3 Radiative forcing by O₃ and CH₄

Radiative forcing (RF) by ozone was approximated using the forcing obtained by the STOCHEM model as described in Dentener et al. (2005), normalized by the ozone columns obtained in that study. Here we use annual averaged forcing based on the RF computations provided as monthly averages by D. Stevenson (personal communication, 2004). The radiative transfer model was based on Edwards and Slingo (1996). These forcings account for stratospheric adjustment, assuming the fixed dynamical heating approximation, which reduces instantaneous forcings by ~22%.

The effect of CH₄ emissions on global forcing is calculated by applying a uniform value of 2.5 mW/m² per Tg CH₄ emitted (Dentener et al., 2010). It includes both the direct CH₄ greenhouse gas (GHG) forcing (1.8 mW/m²) as well as the long-term feedback of CH₄ on hemispheric O₃ (0.7 mW/m²). For CH₄ the RF associated with the base simulation was taken from IPCC-Third Assessment Report (TAR) (Table 6.2 of Ramaswamy et al., 2001).

A secondary feedback through the effect of precursors (NO_x, NMVOC, CO and SO₂) on CH₄, and subsequently on long-term hemispheric O₃ levels, is included as well. The CH₄ response from a perturbation in NO_x, NMVOC and SO₂ is calculated from the change in CH₄ burden due to OH oxidation under the respective perturbations. This burden perturbation is treated as an emission perturbation for CH₄ which translates in a new steady-state CH₄ and background O₃ concentration. Using the HTAP1 simulations SR1 and SR2 (1760 ppb and 1408 ppb, see Dentener et al., 2010) we evaluated a CH₄ lifetime sensitivity coefficient $s = \partial \ln(LT) / \partial \ln(CH_4)$ of 0.33 which can be compared to a range of values between 0.25 and 0.31 in IPCC-TAR (Prather et al., 2001, Table 4.2). The change in steady-state methane concentrations, induced by O₃ precursor emissions, follows the method outlined by Fiore et al. (2008) and Wild and Prather (2000), resulting in a calculated feedback factor $F=1.53$. Fiore et al. (2009) report slightly lower F values of 1.25 to 1.43, due to the fact that they accounted for a fixed CH₄ lifetime with respect to losses to the stratosphere (120 years) and soils (160 years), which would have lowered the TM5 factor F to 1.43, and 5 % higher normalized forcing (Wm⁻²ppb⁻¹). The use of 12 model average F of 1.33 (Fiore et al., 2009) would lead to 12 % higher normalized forcing resulting from indirect O₃ precursor impacts on OH. For simplicity, and given the large uncertainties in soil and stratospheric loss rates, we choose to use the $F=1.53$ factor.

Hence, the greenhouse gas radiative forcing contribution of each ozone precursor consists of 3 components: a direct effect through the production of O₃, a contribution by a change in CH₄ through modified OH levels (including a self-feedback factor accounting for the modified CH₄ lifetime), and a long-term contribution via the feedback of CH₄ on hemispheric ozone.

The response of O₃ forcing to CO emission changes (for which no regional TM5-FASST perturbation model simulations were performed) was taken from TM5-CTM simulations performed for the HTAP1 assessment (Dentener et al., 2010) using the average forcing efficiency for North America, Europe, South-Asia and East-Asia. For regions not covered by the HTAP1



regions, the HTAP1 global average forcing efficiency was used. The resulting region-to-globe emission-based forcing efficiencies are given in Tables S6.2 to S6.5 in the SI for aerosols, CO, CH₄ and other O₃ precursors respectively.

In its current version, TM5-FASST_V0 provides the steady-state concentrations and forcing response of the long-term O₃ and CH₄ feedback of sustained precursor emissions, i.e. it does not include transient computations that take into account the time lag between emission and establishment of the steady-state concentration of the long-term O₃ and CH₄ responses.

2.7.4 Calculation of GWP, GTP, delta T and CO_{2eq} emissions

The obtained emission-based forcing efficiencies are immediately useful for evaluating a set of short-lived climate pollutant climate metrics. Applying the methodology described by Fuglestad et al. (2010) briefly outlined below, the resulting emission-normalized specific forcing responses A_x [W/m²]/[kg/year] are used to calculate the absolute global warming potential (AGWP) and absolute global temperature potential (AGTP) for various time horizons H (20, 50, 100, 500 yr), as a basis to obtain the corresponding CO_{2eq} for the actually emitted amounts.

The AGWP for emitted short-lived (exponentially decaying) species x with lifetime a_x is calculated by integrating the specific forcing over a time span H of an emission pulse at $t=0$:

$$AGWP(H) = \int_0^H A_x \exp\left(\frac{-t}{a_x}\right) dt = A_x a_x \left[1 - \exp\left(\frac{-H}{a_x}\right)\right]$$

AGTP of a short-lived (exponentially decaying) component is calculated as an endpoint change in temperature after H years from a one-year emission pulse at time 0.

$$AGTP(H) = \int_0^H A_x \exp\left(\frac{-t}{a_x}\right) R(H-t) dt$$

Where $R(t)$ represents the response in global-mean surface temperature to a unit pulse in forcing. Following Fuglestad et al. (2010) we adopt the functional form for $R(t)$ from Boucher and Reddy (2008), derived from a GCM :

$$R(t) = \sum_{j=1}^2 \frac{c_j}{d_j} \exp\left(-\frac{t}{d_j}\right)$$

The first term in the summation can crudely be associated with the response of the ocean mixed-layer to a forcing, the second term as the response of the deep ocean with c_j [K(Wm⁻²)⁻¹] and d_j [years] represent temperature sensitivity and response time of both compartments respectively. This leads to:

$$AGTP(H) = \sum_{j=1}^2 \frac{A_x a_x c_j}{(a_x - d_j)} \left(\exp\left(\frac{-H}{a_x}\right) - \exp\left(\frac{-H}{d_j}\right) \right)$$

As discussed earlier, we take into account that species such as NO_x, NMVOC and CO lead to changes in O₃ and CH₄ and consequently have a short-lived component (O₃) as well as long-lived components (CH₄ and CH₄-induced O₃) contributing to AGWP and AGTP. We refer to Appendix 2 in Fuglestad et al., 2010 for a detailed description of the methodology and



numerical values for c_j and d_j . As aerosols and directly produced O_3 from ozone precursors have a lifetime of the order of days (aerosols) to several months (O_3), the resulting integrated specific forcing is insensitive to the actual lifetime for the range of time horizons considered (decades to centuries), and in practice we use a default value of 0.02yr for aerosols and 0.27yr for short term O_3 . This does however not apply to the long-term forcing contribution of CH_4 and the associated O_3 feedback from O_3 precursors for which we use a perturbation adjustment time of 14.2 years (Wild et al., 2001). Note that this adjustment time scale is larger than the total atmospheric time scale for CH_4 oxidation by OH combined with losses to soils and the stratosphere (HTAP1 model ensemble mean: 8.8 years (Fiore et al., 2009)) due to the feedback of CH_4 on atmospheric OH concentrations and thereby its own lifetime (Forster et al., 2007). Fuglestad et al. (2010) report CH_4 adjustment times from various modelling studies between 10.2 and 16.1 years. Dimensionless metrics GWP (GTP) are obtained dividing AGWP (AGTP) by the AGWP (AGTP) of CO_2 as a reference gas for which we use values from Joos et al. (2013).

Finally, still following Fuglestad et al. (2010), we also include a calculation of the global temperature change $\Delta T_x(H)$ between year 0 and year H for a sustained emission change $\Delta E_x(t) = E_x(t) - E_x(0)$ of component x as the sum of the delta T from one-year emission ‘pulses’ approaching the time horizon.

$$\Delta T_x(H) = \sum_{t=0}^H \Delta E_x(t) AGTP(H-t)$$

In this way, a set of climate metrics is obtained which is consistent with the air quality metrics, health and ecosystem impacts calculated from the concentration and deposition fields.

3 Results: validation of the reduced-form TM5-FASST

In this section we focus on validating TM5-FASST_v0, specifically:

- (1) Evaluating possible caveats related to the hypothesis of additivity and linearity of secondary pollutant responses towards single and combined precursor perturbations, by confronting extrapolated FASST SRs (obtained from -20% perturbation) with TM5 simulations for an extended perturbation range [-80%, +100%]
- (2) Testing the validity of the FASST outcome versus TM5 for a set of future emission scenarios that differ significantly from the reference scenario, now including all source and receptor regions in the evaluation.
- (3) Confront FASST key-impact outcomes with results from literature for some selected case studies, with a focus on climate metrics and health impacts.

3.1 Validation against the full TM5 model: additivity and linearity

The standard set of -20% emission perturbation simulations, available for all 56 continental source regions and constituting the kernel of TM5-FASST_v0 are simulations P1 (perturbation of all precursors and primary pollutants emissions), P2 (SO_2 only), and P4 (NH_3 and NMVOC) shown in Table 2. Simulations P3 and P5 (-20%), as well as the additional perturbation



simulations over the range [-80%, +100%] for a limited set of representative source regions (Europe, USA, China, India, Japan) and listed in Annex 4 of the SI, are used to validate the linearized reduced-form approach against the full TM5 model, in particular regarding chemical feedback mechanisms (additivity issue) and extrapolation towards larger emission perturbation magnitudes (linearity issue). This is in particular relevant for the NO_x - NMVOC - O_3 chemistry and for the secondary $\text{PM}_{2.5}$ components NO_3^- - SO_4^{2-} - NH_4^+ . These mechanisms could also be important for organic aerosol, but we remind that in this study organic aerosol formation was parameterized as pseudo-emissions.

3.1.1 Additivity and linearity of secondary inorganic $\text{PM}_{2.5}$ response:

Experiment P1, where BC, POM, SO_2 and NO_x emissions are simultaneously perturbed with -20% compared to base simulation P0, delivers SR matrices for primary components BC and POM, and a first-order approximation for the precursors SO_2 and NO_x whose emissions do not only affect SO_2 and NO_x gas concentrations but also lead to several secondary products (SO_2 forms ammonium sulfate, NO_x leads to O_3 , ammonium nitrate). Experiment P2 perturbs SO_2 only, while experiment P3 perturbs NO_x only (in this latter case, to limit the computational cost, only for a limited set of representative source regions).

We first test the hypothesis that the $\text{PM}_{2.5}$ response to the combined (NO_x + SO_2) perturbation (P1) can be approximated by the sum of the single precursor perturbations responses (P2 + P3). Figure 2 summarizes the results for the selected source regions. Note that for Europe, the emission perturbations were applied over all European countries simultaneously, hence the responses are partly due to inter-regional transport from other countries. Germany as individual source region was evaluated as well, as a case-study with significant emissions and potentially large non-linearities. In general we find that P3 gives a minor response on sulfate, and similar for P2 on nitrate, in general one order of magnitude lower than the direct formation of SO_4^{2-} and NO_3^- from SO_2 and NO_x respectively (Fig. 2). We also find that the -20% perturbations of both precursors behave in an additive manner for what concerns the formation of secondary $\text{PM}_{2.5}$, i.e. $d\text{SO}_4^{2-}/d[\text{SO}_2 + \text{NO}_x] \approx d\text{SO}_4^{2-}/d\text{SO}_2 + d\text{SO}_4^{2-}/d\text{NO}_x$, and similar for NO_3^- and NH_4^+ in $\text{PM}_{2.5}$ (Fig. 2), i.e. $\text{P1} \approx \text{P2} + \text{P3}$. Scatterplots between P1 and P2+P3 for the individual secondary products and total inorganic $\text{PM}_{2.5}$ are shown in Fig. S7.1 of the SI. Hence, from the combined [$\text{SO}_2 + \text{NO}_x$] perturbation (P1), and the separate SO_2 perturbation simulations (P2), which are both available for all source regions, the missing NO_x SR matrices can be gap-filled using ($\text{P1} - \text{P2}$).

Next we evaluate the hypothesis that the -20% perturbation responses can be extrapolated towards larger perturbation ranges as an approximation of a full TM5 simulation. Figure 3 shows, for the selected regions listed in Table S3 of the SI, the TM5 computed change in secondary $\text{PM}_{2.5}$ components, normalized to the base concentration, as a function of the precursor emission perturbation in the range [-80%, +100%] relative to the base emission. The figure illustrates the general near-linear behaviour of responses to single precursor emission perturbations for all regions, except for India where the response linearity towards NO_x emissions breaks down for emission reductions beyond -50%. Linear extrapolation of the -20% responses (i.e. the TM5-FASST approach) towards -80% and +100% perturbations leads to a slight over-prediction of the resulting secondary $\text{PM}_{2.5}$ (i.e. the sum of sulfate, nitrate and ammonium) for all regions considered, in either perturbation



direction (Fig. 4). While the scatter plots for the single perturbations (Fig. 4 a,b,c) evaluate the linearity of the single responses, the plot showing the combined ($\text{SO}_2 + \text{NO}_x$) perturbation (Fig. 4d) is a test for the linearity combined with additivity of FASST. For the -80% perturbation from the base case, the linearized result is within 12%, 19% and 30% of the full TM5 outcome for the single SO_2 , NO_x and NH_3 perturbations respectively, and within 21% of TM5 for the combined ($\text{SO}_2 + \text{NO}_x$) perturbation. The response to a doubling of the base emissions is within 13%, 45% and 12% of the full TM5 outcome for the single SO_2 , NO_x and NH_3 perturbations respectively, and within 34% of TM5 for the combined ($\text{SO}_2 + \text{NO}_x$) perturbation.

The relative errors by region for the [-80%,+100%] perturbation range for individual and combined precursor perturbations versus *total* $\text{PM}_{2.5}$ (primary +secondary) are shown in Fig. S7.2 of the SI, showing that in nearly all cases, the error on total resulting $\text{PM}_{2.5}$ from a simultaneous perturbation on all 3 precursors is higher for a -80% emission reduction than for a doubling of emissions. In nearly all cases, the FASST linearization leads to an over-prediction of total $\text{PM}_{2.5}$, by between 0% and +30%. This information is relevant in view of the general tendency for further emission reduction in developed countries, compared to growing emission in developing regions.

3.1.2 Additivity and linearity of O_3 responses to combined precursor emissions

O_3 atmospheric chemistry is in general highly non-linear, displaying a response magnitude and sign depending on the concentration ratio of its two main ozone precursors NO_x and NMVOC, with high VOC/NO_x ratios corresponding to NO_x -sensitive chemistry and low VOC/NO_x ratios corresponding to VOC-sensitive chemistry (Seinfeld and Pandis, 1998; Sillman, 1999). However a perturbation with simultaneous NO_x and NMVOC emission changes of the same relative size is expected to behave more linearly than single perturbations since the chemical regime remains similar. The FASST reduced-form approach builds on the assumption that the O_3 response to combined precursor perturbation can be approached by the sum of the single component emission perturbations (additivity hypothesis). This is in particular relevant for combined and individual NO_x and NMVOC perturbations, and to a less extend for the (SO_2 , NO_x) combination.

For gap-filling purposes we first evaluate the additivity hypothesis for the combined (SO_2 , NO_x) perturbation. Comparing experiments P1 ($\text{SO}_2 + \text{NO}_x$ perturbation), P2 (SO_2 perturbation) and P3 (NO_x perturbation) confirms that the ozone response to SO_2 emissions is marginal and additive to the response to NO_x ($\text{P1} \approx \text{P2} + \text{P3}$) over the full range of perturbations, as shown in Fig. S7.3 in the SI, and hence we can gap-fill the missing NO_x perturbation SR matrix for all source and receptor regions from ($\text{P1} - \text{P2}$).

Next, we evaluate whether the O_3 response from the combined $\text{NO}_x + \text{NMVOC}$ perturbation (P5) can be approximated by the sum of O_3 responses to individual NO_x (P3) and NMVOC (P4) perturbations, i.e. assuming $\text{P5} = \text{P4} + \text{P3}$. P5 was obtained for a limited set of representative source regions: Europe (by perturbing precursor emissions from all FASST source regions inside the EUR master zoom region simultaneously), China, India and USA. As shown in Fig. 5, also here we find good agreement between the combined ($\text{NO}_x + \text{NMVOC}$) perturbation (open circles) with the sum of the individual



precursor perturbation (black dots). This occurs even in situations where titration by NO_2 causes a reverse response in O_3 concentration as is the case in most of Europa and the USA.

Extending the O_3 (and metrics) linearized responses as a sum of scaled individual -20% precursor responses towards more extreme perturbation ranges could be a challenge, as the individual perturbation of one of the (NO_x , NMVOC) precursor may change the ozone formation regime. In particular during winter time titration of O_3 under high NO_x conditions may reverse the slope of the NO_x emission – O_3 concentration response. However, the impact-relevant O_3 metrics, both health and crop related, are based on summertime and daytime values and are expected to behave more linearly (Wu et al., 2009). Indeed, as shown in Fig. 6, while the response to NMVOC is near-linear and monotonically increasing over the full range for all regions, the NO_x response is showing a more complex behaviour, exhibiting a negative slope for annual mean O_3 over nearly all European regions and the USA while positive for India and China. For the health-relevant exposure metric M6M and the crop metric M12 the slope reverses to positive in most regions, due to their implicit constraint to the summer season when titration plays a minor role, except in strongly NO_x -polluted North-Western European countries (Great Britain, Germany, Belgium and The Netherlands, as well as Finland).

Figure 7 illustrates the performance of TM5-FASST (linearly extrapolated sum of individual responses) versus TM5 for the normalized regional O_3 concentration responses to combined NMVOC and NO_x perturbations for annual mean ozone, M6M, and for the crop-relevant exposure metrics AOT40 and M12 over the extended emission perturbation range. The response (i.e. the *change* between base and perturbed case) to emission reductions down to -80% is generally underestimated by 13% – 17% across the 4 metrics. For a doubling of the combined precursor emissions, the FASST approach over-estimates the response by 20% for annual O_3 , 36% for M12, 50% for M6M and more than a factor 2 for AOT40. Of the four presented metrics, AOT40 is clearly the least robust one, which can be expected for a threshold-based metric that has been linearized. However, for the other metrics even the strong titration effect, leading to reduced O_3 under doubling of NO_x emissions, is still relatively-well reproduced in the reduced-form approach. Table 3 gives the relative error on the resulting estimated metrics (relative to the TM5 outcome) for individual and combined precursor perturbations.

The relative errors by individual region for the evaluated NO_x emission perturbation range for O_3 and O_3 exposure metrics are shown in Fig. S7.4 in the SI. For the selected sample of regions, the relative error on the regional averaged annual ozone falls within [-11%, 8%] (95% CI) of the full model results. The 95% CI for health and crop exposure metrics M6M and M12 are within [1%, 12%] and [-5%, 13%] respectively, while the least robust metric AOT40 falls within [-36%, 95%] of the TM5 outcome, all with respect to NO_x emission perturbations in the range [-80%, +100%]. NMVOC perturbation responses (Fig. S7.5) behave more linearly, with deviations from full model simulations within [0%, 1%], [0%, 3%], [0%, 3%] and [-15%, 20%] for annual O_3 , M6M, M12 and AOT40 respectively. However, North-Western European regions with highest NO_x levels show significantly higher deviations from the full model upon NMVOC perturbations. The errors on FASST NO_x and NMVOC perturbation responses are solely related to the linear extrapolation approach applied and do not include possible errors in the TM5 chemical and transport parameterizations, nor emissions used in the simulations. For the simultaneous NMVOC+ NO_x perturbation, both additivity and linearization errors are introduced (Fig. S7.6). In this case the



95% CI intervals are [-9%, 4%], [1%, 11%], [-2%, 11%], and [-59%, 84%] for annual O₃, M6M, M12 and AOT40 respectively.

3.2 TM5-FASST_v0 versus TM5 for future emission scenarios

In this section we evaluate different combinations of precursor emission changes relative to the base scenario in a global framework. We compare TM5-FASST_v0 results with TM5 output for a set of global emission scenarios which differ significantly in magnitude from the base simulation which used the 2000 emissions, and as such provide a challenge to the application of linear source-receptor relationships used in TM5-FASST. We assume that the full TM5 model provides valid evaluations of emission scenarios, and we test to what extent these simulations can be reproduced by the linear combinations of SRs implemented in the TM5-FASST_v0 model.

We use a set of selected policy scenarios prepared with the MESSAGE integrated assessment model in the frame of the Global Energy Assessment GEA (Rao et al., 2012, 2013; Riahi et al., 2012). These scenarios are the so called “frozen legislation” and “mitigation” emission variants for the year 2030 (named FLE-2030, MIT-2030 respectively), policy variants that describe two different policy assumptions on air pollution until 2030. These scenarios are described in detail in Rao et al. (2013). Major features and emission characteristics are provided in section S8 of the SI. We further note that not only the emission levels of these scenarios are different from the FASST base scenario (RCP year 2000), but also the spatial distribution of the emissions, at the resolution of grid cells, may differ from the reference set.

We compute PM_{2.5} and ozone concentrations applying equation (2), i.e. considering the FLE-2030 and MIT-2030 emission scenarios as a perturbation on the reference emission set (RCP year 2000) from which the TM5-FASST source-receptor matrices were derived. Table S8 in the SI shows (using larger world regions aggregated from the FASST source regions) the change in global emission strengths for the major precursors for both test scenarios, relative to the base scenario on which the SR perturbations were applied.

Emission changes for the selected scenarios mostly exceed the 20% emission perturbation amplitude from which the SRs were derived. Under the MIT-2030 scenario, all precursors and primary pollutants – except NH₃ and primary PM_{2.5} in Asia – are showing a strong decrease compared to the RCP year 2000 reference scenario. The FLE-2030 scenario displays a global increase for all precursors, however with heterogeneous trends across regions, with only Asia undergoing consistently increasing emissions for all pollutants. In other regions (e.g. OECD, Reforming Economies), despite stagnating air pollution controls, emissions decline due to the use of less and cleaner fuels project for 2030.

The MIT-2030 and FLE-2030 emission scenarios were used as input both to TM5-CTM and TM5-FASST_v0. The scope of TM5-FASST is to evaluate on a regional basis the impacts of policies that affect emissions of short-lived air pollutants and their precursors. Hence we average the resulting O₃ and PM_{2.5} concentration and O₃ exposure metric M6M over the each of the 56 FASST regions and compare them with the averaged TM5 results for the same regions. Figure 8 and 9 show the FASST versus TM5 regional scatter plots for PM_{2.5} and annual mean O₃ (as well as M6M) respectively. Despite non-linearities in the response to NO_x and VOC emission changes, regionally aggregated PM_{2.5} and O₃ are reproduced well by



TM5-FASST_v0, nevertheless FASST in general tends to over-estimate $PM_{2.5}$ concentrations, in particular for the low emission case by 54% on the average, 62% for secondary $PM_{2.5}$ and 46% for primary $PM_{2.5}$ (Table 4 and Fig. 8). This is in line with the findings of the limited perturbation experiments described in the previous section. However, in a policy impact analysis framework, the *change* in pollutant concentrations between two scenarios (e.g. between a reference and policy case) is often more relevant than the absolute concentrations. In such a set-up, TM5-FASST_v0 is performing better: while for both high and low emission scenarios FASST over-predicts the absolute $PM_{2.5}$ concentrations, the change in regional $PM_{2.5}$ between the two scenarios is underestimated by 5% on the average, with R^2 of 0.97 (Table 4 and Fig. S8.1).

O_3 metrics (annual mean O_3 concentration and M6M) are, both for the low and high emission scenario cases, slightly over-predicted with the linearized FASST approach, compared to the full TM5 model (Fig. 9). The mean relative deviation between FASST and TM5, as regional averaged metric, evaluated over the results from both scenarios (95% CI) is 2% (-5%, +18%) for annual mean ozone and 10% (1.1%, 34%) for the M6M exposure metric. The change in O_3 and M6M between FLE-2030 and MIT-2030 scenario, in contrast to $PM_{2.5}$, is overestimated compared to TM5 (Table 4 and Figure S8.2) with a zero-offset forced-slope of 1.17 ($R^2 = 0.79$) and 1.04 ($R^2 = 0.82$) between FASST and TM5 for annual mean ozone and M6M respectively.

A major issue in air pollution or policy intervention impact assessments is the impact on human health; therefore we also evaluate the TM5-FASST_v0 outcome on air pollution premature mortalities with the TM5-based outcome, applying the same methodology on both pollution sets. We evaluate mortalities from $PM_{2.5}$ using the IER functions (Burnett et al., 2014) and O_3 mortalities using the log-linear ER functions and RR's from Jerrett et al. (2009) respectively. Figures 10 and 11 show scatterplots of FASST against TM5 mortalities for $PM_{2.5}$ and ozone respectively, as well as total mortalities for major world regions. The latter show that the difference between FASST and TM5 results is smaller than the uncertainty on the mortalities resulting from the uncertainty on RR's only.

3.3 Comparison of TM5-FASST_v0 impact estimates with published studies

In this section we confront TM5-FASST_v0 outcomes for a number of key impacts (climate metrics and human health) with results from earlier studies in the literature.

3.3.1 Year 2000 total global anthropogenic forcing by component

The most widely published radiative forcing estimates compare the present-day with the pre-industrial time. To simulate pre-industrial, for simplicity in our TM5-FASST_v0 evaluation we set all anthropogenic in the base simulation (RCP year 2000) to zero and calculate the change in forcing compared to the base case. We include forcing from all aerosol components, as well as CH_4 (including its feedback on O_3) and the short and long term forcing impacts of NO_x , NMVOC and CO on ozone and the methane lifetime. Figure 12 shows the anthropogenic forcings derived from TM5-FASST by emitted component, together with results from AR5 (year 1750-2011). We find that, except for BC, TM5-FASST_v0 reproduces, within the uncertainties reported by IPCC AR5, the global forcing values by emitted component. Only our estimated BC forcing (0.15



W/m²) falls just outside the AR5 90% confidence interval (0.23, 1.02) W/m², which can be partly explained by the different emission years used in the inter-comparison (also explaining the relatively low estimate for CH₄). However, comparing to another widely used literature source, the TM5-FASST_v0 BC forcing estimate still falls within the 90% CI (0.08, 1.27)W/m² given by Bond et al. (2013) for the year 2005, with a comparable global BC emission rate. Our low-end BC forcing estimate can be partly explained by the simplified treatment as externally mixed aerosol, without accounting for the enhancement of the mass absorption cross-section when BC particles become mixed or coated with scattering components (Bond et al., 2013).

A break-down of the forcing contributions of each emitted pollutant to aerosol, ozone (including immediate and long-term response modes) and methane (when applicable) forcing is given in Table S6.6 of the SI, together with the respective AR5 central values. While there are very large uncertainties associated with the estimates of the indirect aerosol effect due to the strong approximations made in this work, the calculated magnitude (-0.81 W/m²) is in agreement with the published literature range -0.55 W/m² 90% CI (-1.33, -0.06) W/m².

Table 5 compares the contribution of O₃ precursors CH₄, NO_x, NMVOC and CO to the O₃ and CH₄ radiative forcing with earlier work (Shindell et al., 2005, 2009; Stevenson et al., 2013), who used different emission sets than RCP. Our O₃ and CH₄ forcing values from CH₄ and NO_x coincide particularly well with the values obtained by Shindell et al. (2005, 2009) except for the O₃ forcing derived from CH₄ obtained by Shindell et al. (2009) which is 37% higher in their study. In contrast, our estimates of the (less important) CO and NMVOC contributions to O₃ forcing result to be higher by a factor 1.9 and 1.5 for CO and NMVOC respectively compared to Stevenson et al. (2013) and by a factor 2.3 and 7.6 respectively compared to Shindell et al. (2009). The same observation can be made for the contribution of CO and NMVOC to CH₄ forcing, where we find excellent agreement between our results and the two earlier studies by Shindell et al., while our estimates of the contributions from CO and NMVOC are a factor 1.5 and 2.2 higher compared to Stevenson et al. (2013), essentially due to difference in OH levels and lifetimes across models.

3.3.2 Direct radiative forcing of short-lived climate pollutants by sector

The segregation of the RCP reference emission inventory by sector enables the evaluation of the contribution of individual sectors to the global instantaneous forcing. This is achieved by ‘switching off’ the respective sectorial emissions in the base emission scenario one by one, and comparing the resulting ΔForcing with the reference case. We compare the total and sector-attributed direct radiative forcing with Unger et al. (2010) who made a similar evaluation for the year 2000 based on the EDGAR Fast Track 2000 emission inventory (Olivier et al., 2005). Figure 13b shows the break-down by forcing component (where we separated the direct and indirect contributions to O₃ and CH₄ forcing), while the Fig. 13a shows the contributions by emission sector. Since different inventories are used, we do not expect a perfect match between the two analyses, however the emerging picture, in terms of over-all contribution by emitted component, as well as the contribution by sector is very similar, underlining the applicability of the TM5-FASST tool for this type of analysis in a consistent framework with other types of impacts. In general, BC forcing as well as the short-term O₃ forcing by NO_x and NMVOC



(SLS-O₃) are consistently lower for FASST, while the indirect feedbacks on CH₄ and long-term O₃ are corresponding well. This is also the case for the direct forcing by inorganic aerosols and POM. The higher direct CH₄ forcing and its feedback on O₃ by Unger et al. (2010) can be attributed to higher emissions in particular in the agricultural and waste – landfills sectors.

5 3.3.3 GWP and GTP

We use the methodology described in section 2.7.4 to evaluate global GTP and GWP for different time horizons H (20y and 100y) and compare with the range of values given in IPCC AR5 (Myhre et al., 2013). We recall that the forcings used to compute the FASST metrics, based on the meteorological year 2001 and RCP year 2000 emissions, are region-specific and take into account differences in atmospheric life time and surface albedo. As shown in Table 6 we find an over-all good agreement with AR5 values. TM5-FASST BC metrics are at the low end of the IPCC range, in line with the previously made observation regarding the low FASST BC forcing. For the NO_x metrics we have separately reported the strongly different ranges from Fuglestad et al. (2010) and Shindell et al. (2009). Our values for NO_x appear to be more in line with the former study, except for GWP20 where FASST gives a negative value (-31) and whereas AR5 reports a range (12, 26) from Fuglestad et al. (2010) and (-440, -220) from Shindell et al. (2009).

15 3.3.4 Health impacts: intercomparison with ACCMIP model ensemble

The health impact analysis of the RCP scenarios performed with the Atmospheric Chemistry and Climate Model Intercomparison Project (ACCMIP) model-ensemble (Silva et al., 2016), provides a useful test case for the ability of TM5-FASST to reproduce trends derived from emission scenarios. The ACCMIP ensemble consisted of 14 state-of-the-art global chemistry climate models with spatial resolution from 1.9°x1.2° to 5°x5°. The ACCMIP models simulated future air quality for specific periods through 2100, for four global greenhouse gas and air pollutant emission scenarios projected in the Representative Concentration Pathways (RCPs). The analysis by Silva et al. (2016) used the same methodology as implemented in FASST for estimating premature mortalities from PM_{2.5} and O₃ as in TM5-FASST (i.e. Burnett et al., 2014; Jerrett et al., 2009 respectively), with the small difference that it does not include Acute Lower Respiratory Infections (ALRI) as death cause (in FASST applicable to age group below 5 years only) and the evaluated age group is >25 years old while in TM5-FASST this was done for population older than 30 years. Further, the ACCMIP health impact analysis uses scenario-specific projections for population and cause-specific base mortalities while FASST uses the same population projections and mortality rates as described in the methods section across all scenarios.

Following the approach of Silva et al. (2016), we compare the global population-weighted annual mean PM_{2.5} concentration change and ozone exposure metric M6M relative to year 2000 concentrations for RCP scenarios 2.6, 4.5 and 8.5 for the years 2030 and 2050, with year 2000 exposure evaluated over the population of the respective scenario years (Tables S2 and S3 in Silva et al., 2016). Figure 14 shows the results from the ACCMIP model ensemble as well as individual model results along with TM5-FASST outcome. We make the evaluation with and without the urban increment parameterization included (using



the generic increment factors from Table S4.2). We find that TM5-FASST qualitatively reproduces $PM_{2.5}$ trends (i.e. the slope) between 2030 and 2050 for the selected RCP scenarios although in only 2 of the 6 considered scenarios the TM5-FASST concentration relative to year 2000 falls within the ACCMIP ensemble range. Even without urban increment correction, TM5-FASST consistently gives higher $PM_{2.5}$ exposure levels than ACCMIP (higher by 0.9, 1.5 and 1.0 $\mu g m^{-3}$ in 2030 and 0.7, 1.3 and 0.9 $\mu g m^{-3}$ in 2050 for RCP 2.6, 4.5 and 8.5 respectively). A plausible explanation is the underlying higher spatial resolution in FASST ($1^\circ \times 1^\circ$) than any of the ACCMIP models. Including the urban increment increases the global mean change in exposure relative to year 2000 with an additional 0.1 to 0.6 $\mu g m^{-3}$.

The ozone exposure metric M6M falls within the range of the ACCMIP model ensemble for 2030 - 2050, but the slope between 2030 and 2050 is lower than for the ACCMIP ensemble mean. In the light of the previously demonstrated satisfactory performance of FASST versus TM5, this indicates that inter-model variability is a stronger factor in the model uncertainty than the reduced-form approach.

The trends from 2000 to 2050 in global mortality burden from $PM_{2.5}$ and O_3 are shown in Figure 15. Assuming that the relative error for the year 2000 – the only uncertainty range given by Silva et al. (2016) – can be applied on the other cases, we find that TM5-FASST reproduces the ACCMIP health impacts from $PM_{2.5}$ within the ACCMIP range. Including the urban increment correction increases the mortality by 26% in 2000, 24%, 22% and 17% in 2030, and 32%, 31% and 25% in 2050 for RCP2.6, 4.5 and 8.5 respectively.

While also O_3 mortalities for years 2000 and 2030 are within the ACCMIP range, TM5-FASST does not confirm the strongly increasing O_3 mortalities in the ACCMIP ensemble by 2050. However this difference can be to a large extent attributed to the use of different baseline mortality and population statistics, in particular for the year 2050 where FASST, by lack of WHO projections for 2050, assumes year 2030 WHO projected mortality rates whereas Silva et al. (2016) use International Futures (IFs) projections up till 2100. Indeed, the IFs projections (Fig. S7 in the SI of Silva et al., 2016) foresee relative constant global mortality rates (deaths per 1000 people) between 2030 and 2050 for all air pollution-related death causes, except for respiratory disease (on which O_3 mortality estimates are based) which increases with a factor 2.5 globally from 2030 to 2050. An acceptable agreement with the ACCMIP model ensemble outcome is achieved when this effect is included as a simple adjustment factor on the FASST RCP year 2050 O_3 mortalities, as shown by the dot-symbols (year 2050) in Fig. 15.

A regional break-down of mortality burden from $PM_{2.5}$ in 2030 and 2050, relative to exposure to year 2000 concentrations, for major world regions and for the globe is shown in Figure S9. The results demonstrate that FASST has the capacity to deliver the essential regional features. In line with the conclusions drawn from the comparison with the full TM5 model, the results in Figure S9 confirm that FASST tends to under-predict the benefit of emission reductions, while over-predicting the impact of increasing pollution.



4 Discussion

Although the methodology of a reduced-form air quality model, based on linearized emission – concentration sensitivities is not new and has been successfully applied in earlier studies (Alcamo et al., 1990), the concept of directly linking pollutant emission scenarios to a large set of impacts across various policy fields, in a global framework, have made TM5-FASST a highly requested tool in a broad field of applications. The results in the previous sections have outlined its strengths and weaknesses. The major strength of the tool is its mathematical simplicity allowing for a quick processing of large sets of scenarios or scenario ensembles. An extreme example is the full family of SSP scenarios delivered by all participating Integrated Assessment Models, for decadal time slices up to 2050, constituting a batch of 594 scenarios of which a selection of 124 scenarios was analyzed with TM5-FASST in the study by Rao et al. (2017). Further, the tool is unique in having a broad portfolio of implemented impact modules which are evaluated consistently over the global domain from the same underlying pollutant field which creates a basis for a balanced evaluation of trade-offs and benefits attached to policy options.

On the other hand, the reduced-form approach inevitably encompasses a number of caveats and uncertainties that have to be considered with care. The reliability of the model output in terms of impacts depends critically on the validity of the linearity assumption for the relevant exposure metrics (in particular secondary components), which becomes an issue when evaluating emission scenarios that deviate strongly from the base and 20% perturbation simulations from which the SR have been established. The previous sections have shown that when aggregated at the regional level, non-linearity effects in PM_{2.5} and O₃ metrics remain within acceptable limits. The available extended-range perturbation simulations could form the basis of a more sophisticated parameterization including second order terms following the approach by Wild et al. (2012) both for O₃ and secondary PM_{2.5}.

Another issue for caution relates to the spatial distribution emissions for arbitrary scenarios analysed with FASST. Having established the SR matrices ‘once and for all’, the underlying emissions spatial distribution patterns are implicitly fixed within each source region. In other words, for the analysis of arbitrary emission scenarios of the past or the future, the FASST model will scale the emissions in each grid cell by the ratio of total region scenario emissions to year 2000 emissions for that region, but it cannot deal with shifting emission patterns of a specific compound compared to the base simulation year 2000 *within* a single source region. In practice this is not expected to introduce large errors as anthropogenic emissions are closely linked to populated areas and road networks of which the extent may be change, but much less so the spatial distribution. It can be a problem when going far back in time, when large patterns of migration and land development occurred, while in RCP scenarios relatively simple expansions of emissions into the future did not assume huge shifts in regional emission patterns. It can be expected that newer generation scenarios with dynamic allocation of emission across countries and macro-regions, for which FASST errors will be larger.

The implicitly fixed emission spatial distribution is also relevant when making a sector apportionment of pollutant concentrations and impacts. Source-Receptor relations are indeed particularly useful to evaluate the apportionment of emission sources (in terms of economic sector as well as source regions) to pollutant levels in a given receptor. However, as



the TM5-FASST_v0 source-receptor matrices were not segregated according to economic sectors, an emission reduction of 20% for a given source region is implicitly considered as a 20% reduction in all sectors simultaneously. While the atmospheric chemistry and transport of emissions is in principle independent of the specific source, a difference in the sector-specific SR matrices may occur due to differences in temporal and spatial (horizontal/vertical) distribution of the sources. Therefore apportionment studies on sectors which have a significantly different emission spatial distribution than other sectors in the same region should be interpreted with care. In particular impacts of off-shore flaring cannot be assessed with TM5-FASST because those emissions were not included in the RCP base emissions.

Comparing to earlier studies and reference data, the performance of TM5-FASST with respect to climate metrics is satisfactory, with the exception of BC forcing which is at the low side of current best estimates. In fact, earlier TM5-FASST assessments where climate metrics were provided (UNEP, 2011; UNEP and CCAC, 2016) applied an adjustment factor of 3.6 on BC forcing, in line with the observation by Bond et al. (2013) that many models underestimate atmospheric absorption attributable to BC with a factor of almost 3. In TM5 FASST, an adjustment factor of 3.6 leads to a global forcing by anthropogenic BC of 600 mW m^{-2} .

Ozone impact on agricultural crop production is deemed to be the least robustly quantified impact category included in FASST, in particular when evaluated from the threshold-based AOT40 metric, and has to be interpreted as indicative order-of-magnitude estimate. In an integrated assessment perspective of evaluating trade-offs and benefits of air pollutants scenarios, the dominant impact category however appears to be human health (Kitous et al., 2017; OECD, 2016; UNEP, 2011) where TM5-FASST provides reliable estimates.

5 Conclusions and way forward

The FASST_v0 version of TM5 is a trade-off between accuracy and applicability. TM5-FASST_v0 enables immediate “what-if” and sensitivity calculations, and, by means of the available source-receptor coefficients, the extraction of this information down to the level of individual regions, economic sectors and chemical compounds. In this paper we have extensively documented the embedded methodology and validated the tool against the full chemistry transport model as well as against selected case studies from the literature. In conclusion, provided that the TM5-FASST_v0 is considered as a screening tool, the simplifications introduced in order to generate immediate results from emission scenarios are not compromising the validity of the output and as such TM5-FASST_v0 has been proven to be a useful tool in science-policy analysis.

The native set of TM5-FASST region-to-grid source-receptor grid maps is sufficiently detailed, both in terms of spatial and temporal resolution as well as number of pollutant species and metrics, to include additional impact categories not included so far. Some examples are BC deposition to snow-covered surfaces, combined nitrogen fertilization and O_3 feedbacks on Carbon-sequestration by vegetation from NO_x emission, both relevant as additional climate forcing, population exposure to NO_2 and SO_2 as additional health effects, ...



The regional 58x56 region-to-region source-receptor matrices aggregated from the high-resolution (region-to-gridmap) SRs are easily implemented in a spreadsheet-type environment. A user-friendly web-based interactive version based on the latter is available at <http://tm5-fasst.jrc.ec.europa.eu/>.

Some foreseen further developments of the TM5-FASST tool, making use of readily available SRs include:

- 5 – Using the available extended-range perturbation simulations to develop a correction algorithm on the current simple linear extrapolation procedure, in particular for the regions where the O₃ or secondary PM_{2.5} regimes are non-linear, e.g. following the approach by Wild et al (2010) and Turnock et al. (2018)
- Update the health impact modules with recent findings in literature, specifically on the long-term O₃ impact (Turner et al., 2016), adjusted IER function parameters and age-specific exposure – response functions for PM_{2.5}
- 10 mortalities (Cohen et al., 2017), as well as including different health metrics (DALYS, life years lost) and improved projections for base mortalities and other health statistics.
- Including a transient O₃ response function to CH₄ emission changes
- Including cryosphere forcing via BC deposition
- Stomatal approach for crop ozone impacts and extension of vegetation types considered
- 15 – Higher temporal resolution exploiting the available native monthly source-receptor maps.

Even with these further developments, an important limitation of TM5-FASST_v0 remains that it is based on a single meteorological year (2001), on source-receptor relations computed by a single underlying Chemistry-Transport model, based on the reference year 2000, and using fixed fields for natural PM_{2.5}. The HTAP phase 2 modelling exercise addresses these

20 issues: it has been designed in line with the FASST philosophy (albeit with a larger aggregation of source region definitions), with an *ensemble* of chemistry-transport or climate-chemistry models providing source-receptor simulations, based on an updated and harmonized common anthropogenic pollutant emission inventory for the years 2008 - 2010 (Janssens-Maenhout et al., 2015; Koffi et al., 2016). Efforts are now underway to implement the HTAP2 ensemble of source-receptor relations in the FASST architecture, resulting in a new web-based and user-friendly HTAP-FASST version, thus creating a link between

25 the knowledge generated by the HTAP scientific community and interested policy-oriented users.

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Table 1: Relevant precursor-pollutant relationships included in TM5-FASST. ●: direct emission or immediate product; ○: effect via thermodynamic equilibration; ◇ effect via first order oxidation products (OH) affecting the lifetime of other precursors.

Pollutant Precursor	SO ₂ (g)	NO _x (g)	NH ₃ (g)	O ₃	CH ₄	SO ₄ (PM _{2.5})	NO ₃ (PM _{2.5})	NH ₄ (PM _{2.5})	EC (PM _{2.5})	POM (PM _{2.5})	SO _x (dep)	NO _y (dep)	Rad. forcing
SO ₂ (g)	●	◇	○	◇	◇	●	○	○			●		●
NO _x (g)	◇	●	○	●	◇	○	●	○			◇	●	●
NH ₃ (g)	◇	◇	●	◇	◇	○	○	●			◇		●
BC (pm)									●				●
POM (pm)										●			●
NMVOC (g)	◇	◇	◇	●	◇	◇	◇	◇			◇		●
CO (g)*				●	◇								●
CH ₄ (g)*	◇	◇	◇	◇	●	◇	◇	◇			◇		●

* From HTAP phase 1 (Dentener et al., 2010)



Table 2: Overview of TM5-CTM perturbation simulations (20% emission reduction) for the calculation of the source-receptor (SR) matrices*comparing to the same zoom regions as in P0.

Simulation	Emission perturbations	Applied on source regions	Scope
P0	No perturbations	Master zoom regions with 1°x1° resolution: AFR, AUS, EAS, EUR, MAM, MEA, NAM, RSA, RUS, SAM, SAS, SEA and PAC (3°x2°)	Base simulation
P1	SO ₂ , NO _x , BC, POM	All 56 continental regions* + international shipping + aviation	SR matrices for BC and POM and first order approximation for SO ₂ and NO _x , assuming negligible chemical interaction
P2	SO ₂	All 56 source regions* + shipping	Independent SR for SO ₂ , to be compared to P1 to quantify potential interference between SO ₂ and NO _x in the formation of sulfate and ozone
P3	NO _x	Representative source regions* (China, Europe, Japan, India, Germany, South-Africa, USA)	Independent SR for NO _x , to verify the additivity of P1 = P2 + P3 and justify the use of (P1 – P2) as a proxy for NO _x perturbation for all other regions
P4	NH ₃ , NMVOC	All 56 continental source* regions + international shipping	SR matrices for NH ₃ and NMVOC emissions, assuming little chemical interaction between the selected precursors in the formation of NH ₄ and O ₃
P5	NMVOC, NO _x	Representative source regions* (Europe, China, India, USA)	Quantify chemical feedbacks in O ₃ formation between NO _x and NMVOC (P5 = P3 + P4) additivity

*See list of regions and their definition in Table S2.2 of the SI.



Table 3: Median (95% CI) relative error on annual O₃ and O₃ metrics (regional averages) from TM5-FASST_v0 versus TM5, for the set of selected regions listed in Table S3 of the SI. M12 and AOT40 are obtained from grid cell's maximal 3-monthly value observed during the year.

Precursor	PERTURBATION	$\frac{C^{FASST} - C^{TM5}}{C^{TM5}} (\%)$			
		Annual O ₃	M6M	M12	AOT40
NO _x	-80%	5.0 (-0.6, 10)	5.8 (2.6, 12)	4.9 (1.0, 9.1)	30 (5.5, 84)
	100%	2.2 (-11, 8.0)	5.6 (1.1, 9.5)	4.3 (0.0, 11)	16 (0.4, 47)
NMVOC	-80%	0.4 (0.2, 1.6)	0.6 (0.2, 2.8)	0.5 (0.1, 1.3)	2.3 (-7.5, 6.5)
	100%	0.8 (0.4, 2.3)	1.1 (0.4, 3.6)	1.0 (0.1, 1.9)	3.3 (-3.4, 7.3)
NO _x +NMVOC	-80%	2.4 (-2.2, 5.4)	2.5 (-1.0, 4.4)	2.1 (0.3, 4.4)	28 (0.9, 69)
	100%	1.2 (-8.9, 4.9)	4.1 (1.2, 12)	3.9 (-0.5, 6.9)	28 (16, 51)



Table 4: Linear regression parameters (forced to zero offset) for regional population-weighted mean pollutant concentrations computed with TM5-FASST-V0 versus TM5-CTM, for two emission scenarios FLE-2030 and MIT-2030 (see text) and for the delta concentration between the two scenarios

	FLE		MIT	
	Slope	R ²	Slope	R ²
Total PM _{2.5}	1.11	0.97	1.54	0.85
Primary PM _{2.5}	1.20	0.96	1.62	0.91
Secondary PM _{2.5}	0.93	0.95	1.46	0.42
Annual mean O ₃	1.04	0.90	1.03	0.92
M6M	1.09	0.90	1.13	0.86

	Slope Delta (FLE – MIT)	R ² Delta (FLE – MIT)
Total PM _{2.5}	0.95	0.97
Primary PM _{2.5}	1.04	0.96
Secondary PM _{2.5}	0.76	0.94
Annual mean O ₃	1.17	0.79
M6M	1.04	0.82



Table 5: Contributions of emissions of CH₄, NO_x, CO and NMVOC to O₃ and CH₄ radiative forcing. Stevenson et al. (2013): for the period 1850-2000; Shindell et al. (2005, 2009) for the period 1750-2000. FASST: anthropogenic emissions RCP year 2000

	Stevenson et al., 2013	Shindell et al., 2005	Shindell et al., 2009	TM5-FASST
Contribution to O ₃ forcing (mWm ⁻²)				
CH ₄	166 ± 46	200 ± 40	275	200
NO _x	119 ± 33	60 ± 30	41	61
CO	58 ± 13		48	108
NMVOC	35 ± 9		7	53
Contribution to CH ₄ forcing (mWm ⁻²)				
CH ₄	533 ± 39	590 ± 120	530	500
NO _x	-312 ± 67	-170 ± 85	-130	-167
CO	57 ± 9			83
NMVOC	22 ± 18			49



Table 6: Global GWP and GTP values 95% CI range (excluding Indirect Radiative Effects) from IPCC AR5 (Forster et al., 2007), and from FASST based on RCP year 2000 emissions and the regional forcing efficiencies listed in Table A6.2 of the SI (all numbers rounded to 2 significant figures).

	GWP20		GWP100		GTP20		GTP100	
	AR5	FASST	AR5	FASST	AR5	FASST	AR5	FASST
CH ₄	(70, 98)	78	(24, 33)	29	(56, 79)	66	(3.6, 5.0)	3.9
BC	(940, 4100)	880	(257, 1100)	240	(270, 1200)	340	(35, 150)	37
OC	(-410, -89)	-280	(-114, -25)	-77	(-120, -26)	-110	(-16, -3)	-12
SO ₂	(-210, -70)	-150	(-58, -19)	-40	(-61, -20)	-57	(-8, 38)	-6.2
VOC	(8.3, 20)	21	(2.7, 6.3)	7	(4.4, 11)	11	(0.4, 0.9)	1.2
NO _x	(12, 26) ^a	-31	(-15, -7) ^a	-14	(-120, -57)	-100	(-3.9, -1.9)	-8
	(-220, -440) ^b		(-130, -64) ^b					
CO	(6.0, 7.8)	7.9	(2, 3)	2.6	(3.7, 6.1)	6.3	(0.27, 0.55)	0.42

a Fuglestvedt et al. (2010)

b Shindell et al. (2009), excluding indirect aerosol effects

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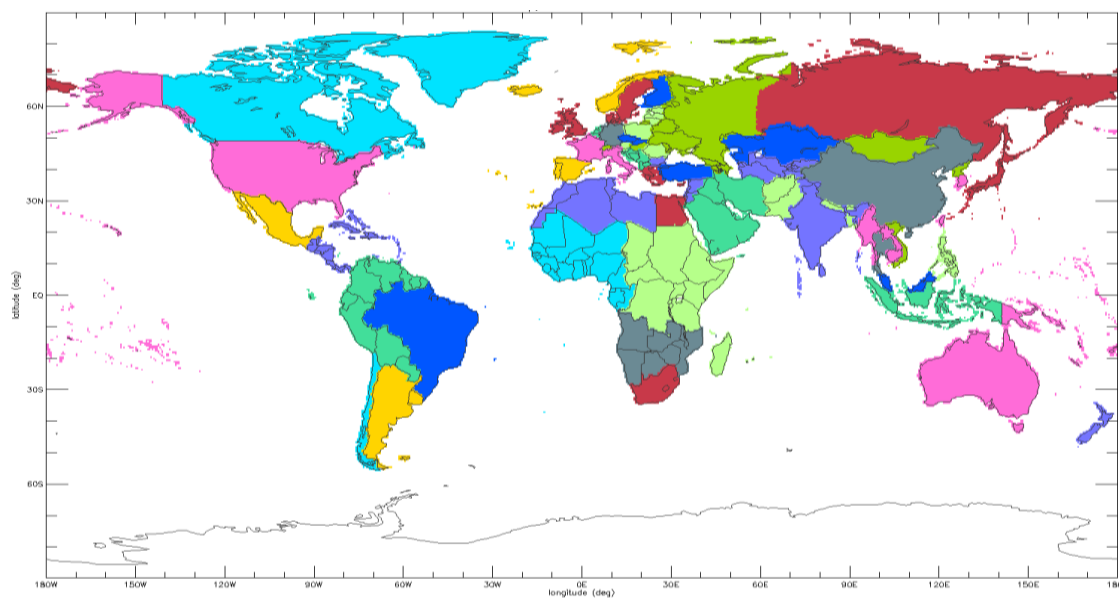


Figure 1: 56 continental emission source regions in TM5-FASST. See Table S2.2 in the SI for the mapping between regions and countries

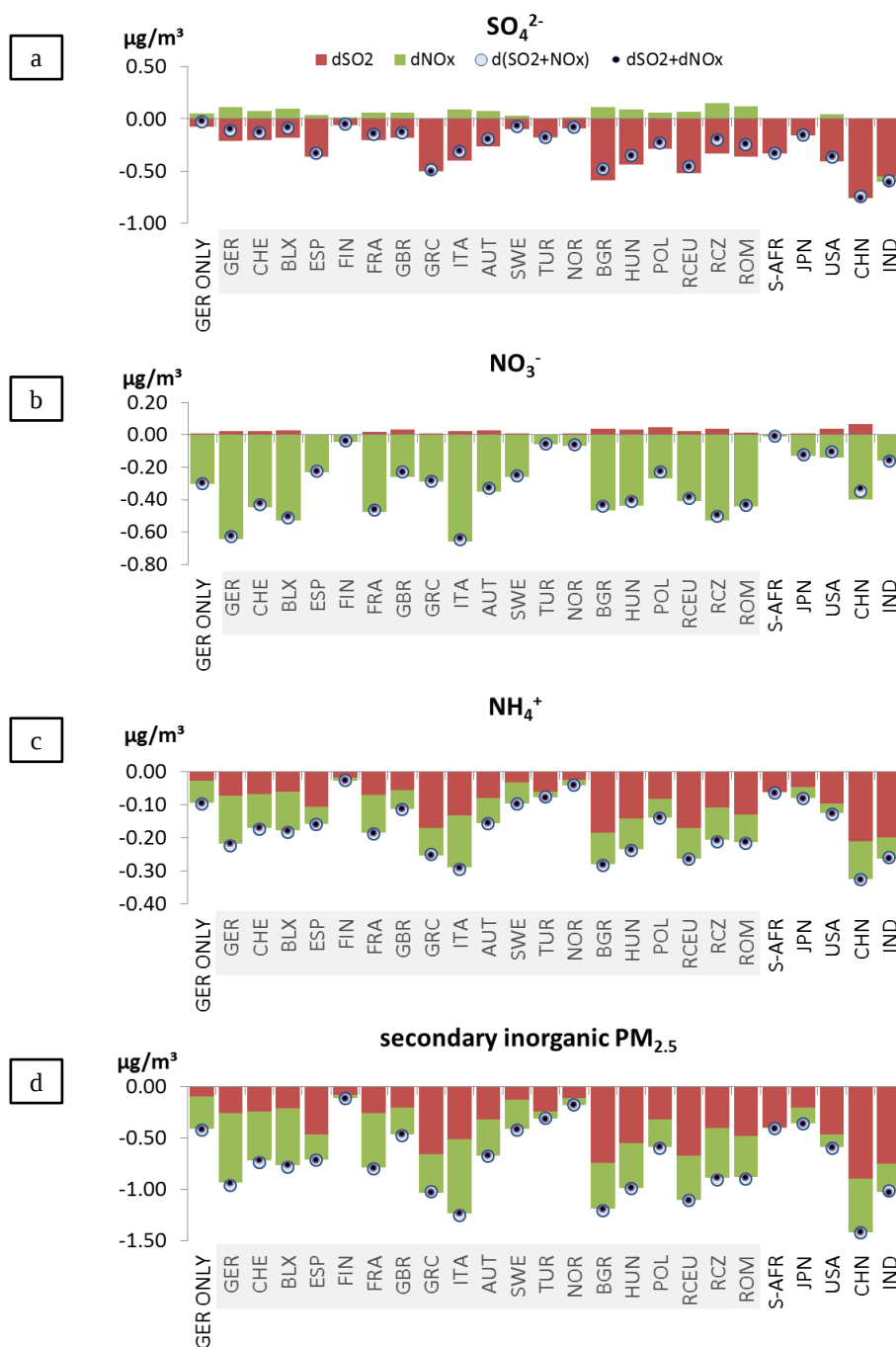


Figure 2: TM5-CTM response in annual population-weighted mean sulfate (a), nitrate (b), ammonium (c) and total inorganic secondary $\text{PM}_{2.5}$ (d) (as sum of the 3 components) upon emitted precursor perturbation of -20% for selected source regions (see SI table S2.2 for the region codes legend). Only the concentration change inside each source region is shown. Red bars: SO_2 -only perturbation (simulation P2); green bars: NO_x -only perturbation (simulation P3). Open circles: simultaneous ($\text{SO}_2 + \text{NO}_x$) perturbation (simulation P1). Black dots: P2 + P3. Shaded regions are perturbed simultaneously as one European region.

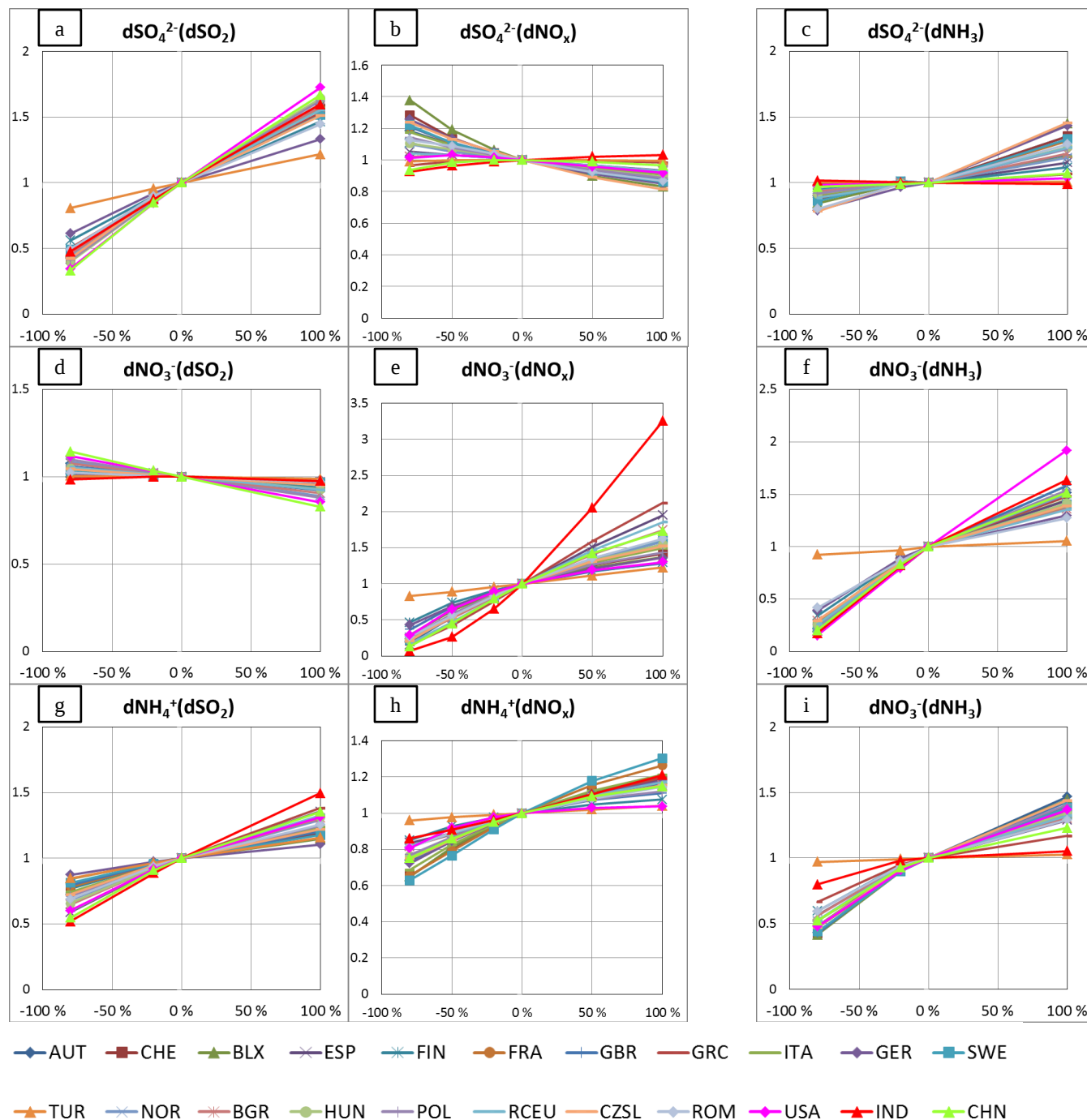


Figure 3: TM5-CTM change in population-weighted regional mean secondary PM_{2.5} components SO_4^{2-} (a to c), NO_3^- (d to f), NH_4^+ (g to i), relative to their respective base scenario concentration, as a function of precursor SO_2 (a, d, g), NO_x (b, e, h) and NH_3 (c, f, i) emission perturbation strength for European receptor regions, USA, India and China. Perturbations were applied over all European regions simultaneously.



PM_{2.5} linearity of SO₂, NO_x and NH₃ response

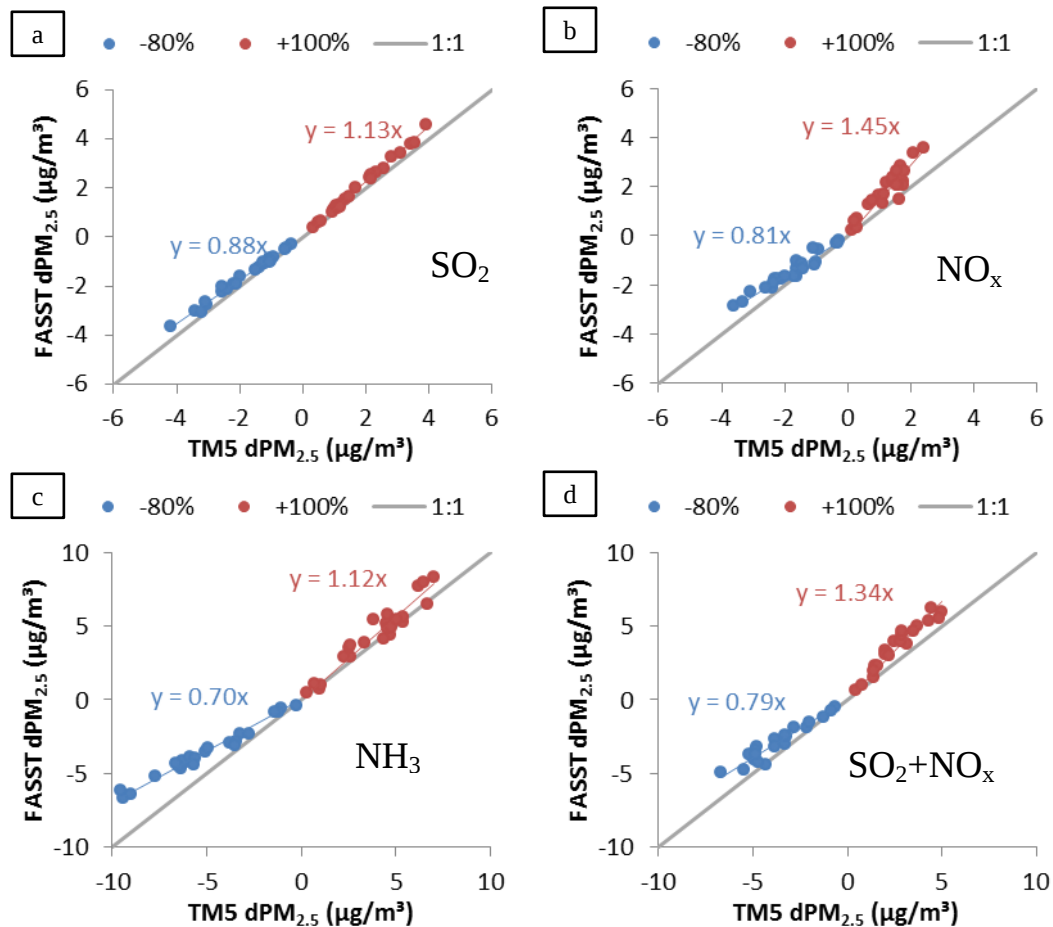


Figure 4: Regional Secondary PM_{2.5}(SO₄²⁻+NO₃⁻+NH₄⁺) response to -80% and +100% single precursor emission perturbations for SO₂ (a), NO_x (b), NH₃ (c) as well as the combined SO₂ + NO_x perturbation (d). X-axis: Full TM5 model; Y-axis: Linear extrapolation of -20% perturbation (FASST approach). Each point corresponds to the population-weighted mean concentrations over a receptor region.

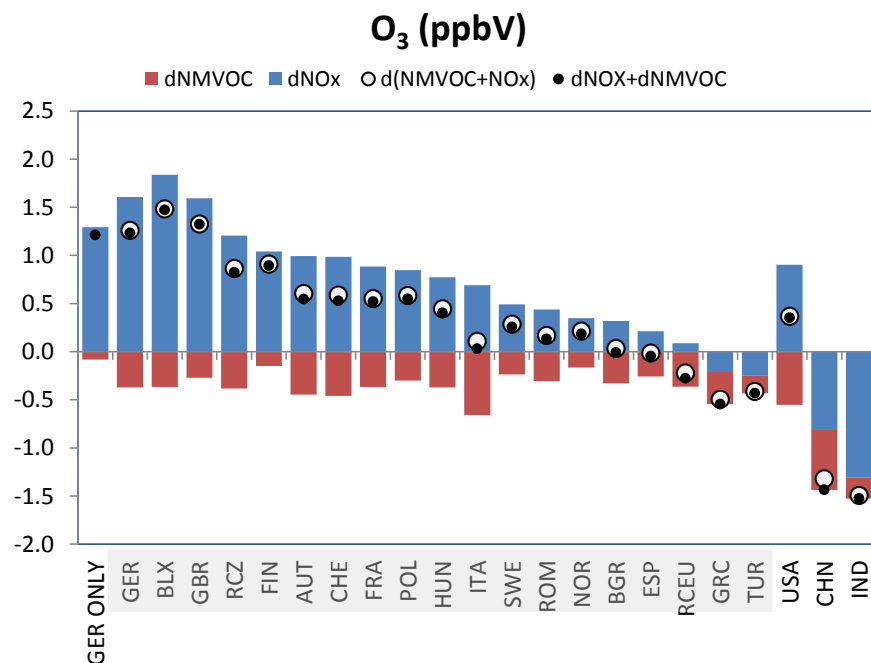


Figure 5: TM5-CTM response in annual mean population-weighted O₃ concentration (in ppbV) upon emitted precursor perturbation of -20% for selected source receptor regions. European regions were perturbed simultaneously. Red bar: NMVOC-only perturbation (simulation P4); blue bar: NO_x-only perturbation (simulation P3). Open circles: simultaneous (NMVOC + NO_x) perturbation (simulation P5). Black dots: P3 + P4. Shaded regions are perturbed simultaneously as one European region.

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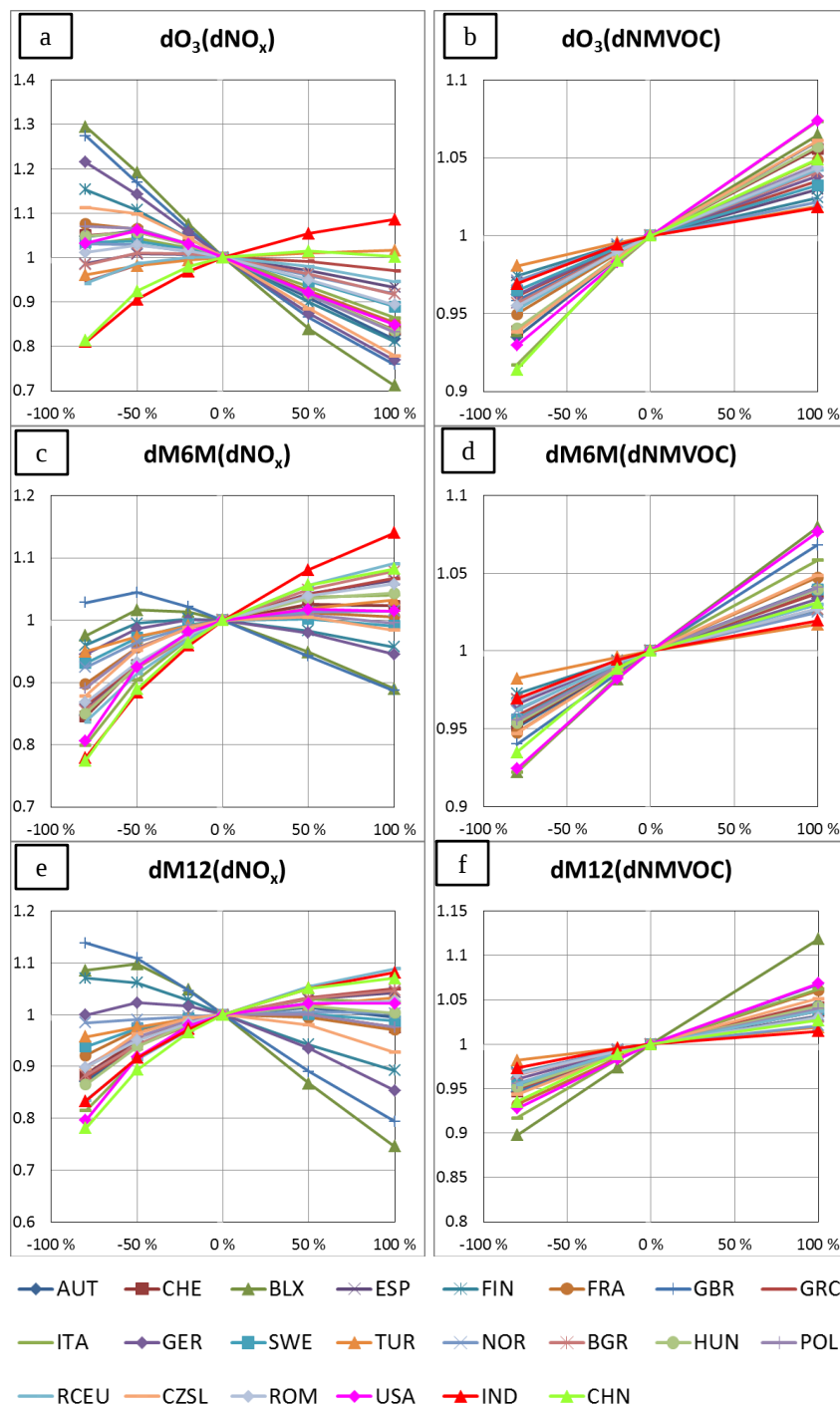


Figure 6: TM5-CTM response in population weighted annual mean O_3 (a, b) and health exposure metric M6M (c, d), and in grid cell-area-weighted crop exposure metric M12 (e, f), relative to their respective base simulation values, as a function of precursors NO_x (a, c, e) and NMVOC (b, d, f) emission perturbation strength. European regions are perturbed simultaneously as one region.

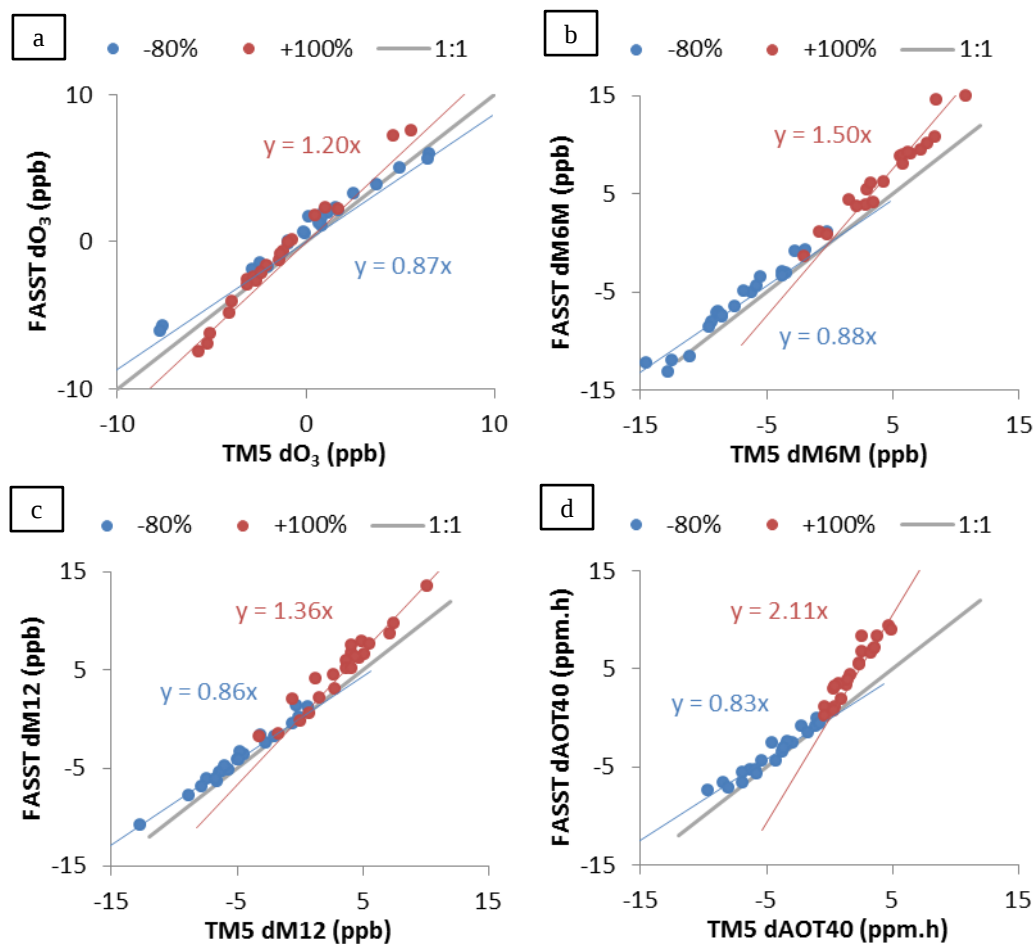


Figure 7: Regional O_3 and O_3 exposure metrics responses to combined -80% and +100% precursor emission perturbations of NO_x and NMVOC. (a) annual mean population-weighted O_3 ; (b) population-weighted M6M; (c) area-mean M12; (d) area-mean AOT40 X-axis: Full TM5 model; Y-axis: Linear extrapolation of -20% perturbation (FASST approach). Each point corresponds to the mean metric over a source region.

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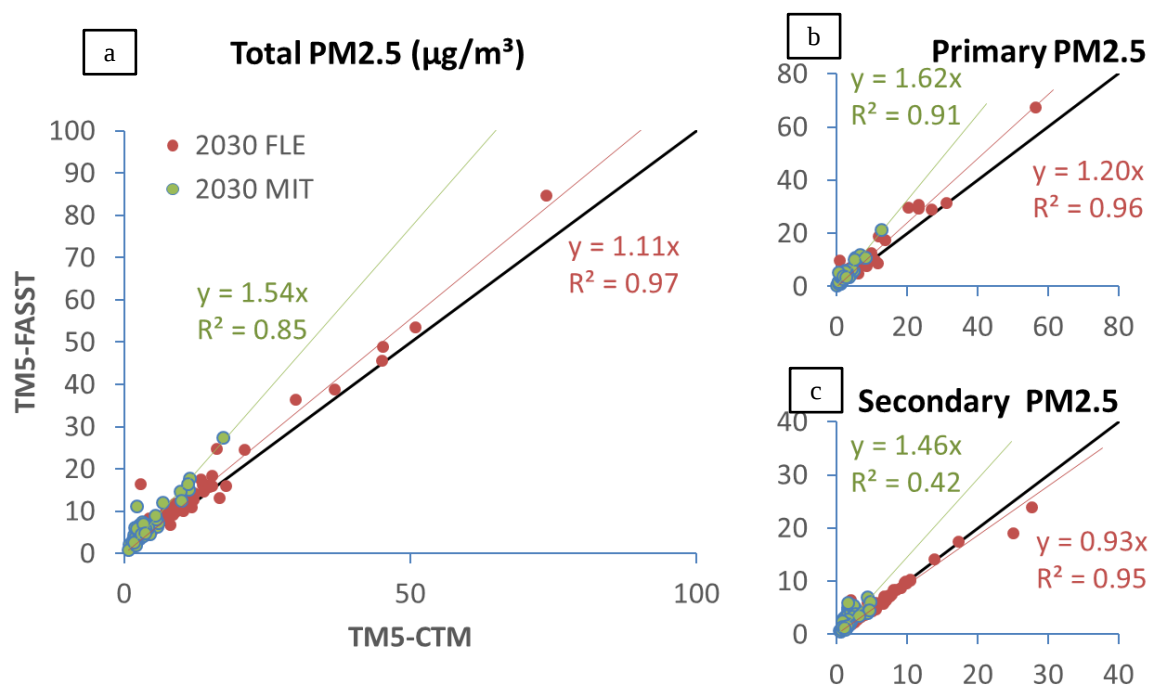


Figure 8: (a) PM_{2.5} concentration obtained with TM5-FASST versus TM5-CTM for high (FLE, red dots) and low (MIT, green dots) emission scenarios (see text). Each point represents the population-weighted mean over a TM5-FASST receptor region. Black line: 1:1 relation. breakdown for (b) primary (BC+POM+other primary PM_{2.5}) and (c) secondary (SO₄+NO₃+NH₄) PM components (same axis definitions as left plot).

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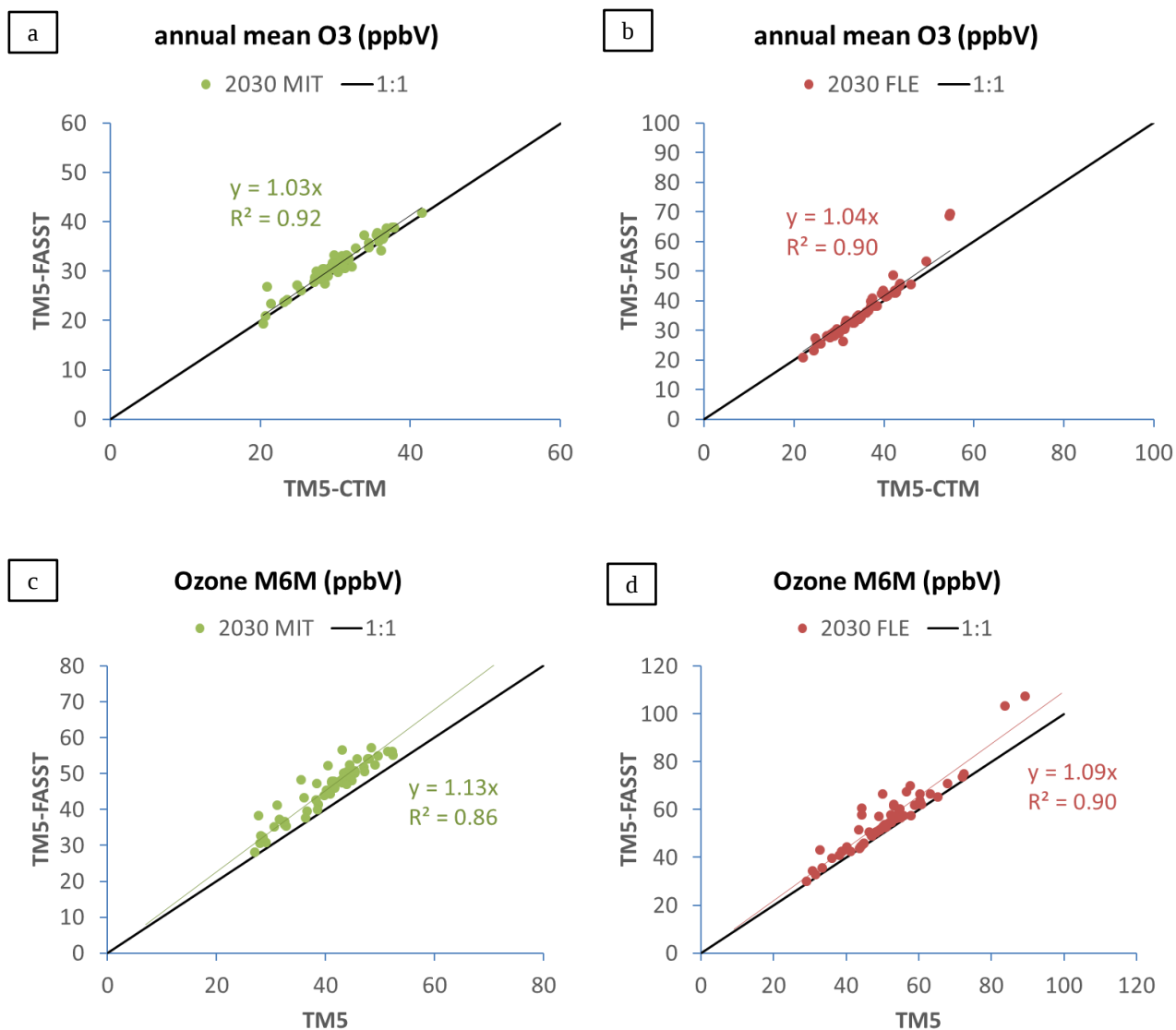


Figure 9: Scatter plot of population-weighted mean ozone (a, b) and exposure metric M6M (c, d) obtained by TM5-FASST versus TM5 chemistry transport model (CTM), for a low (MIT, left) and high (FLE, right) emission scenario. Each point represents the population-weighted mean over a TM5-FASST receptor region



Premature mortalities from PM_{2.5}

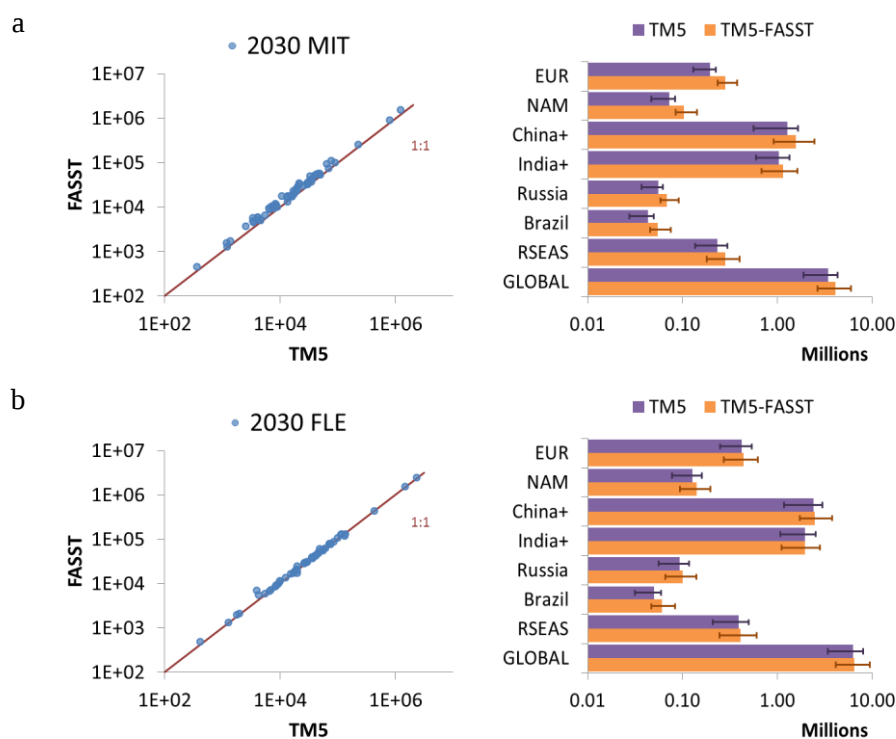


Figure 10: FASST versus TM5 premature mortalities from exposure to PM_{2.5} for MIT(a) and FLE-2030 (b) scenarios (see text). Dots: aggregated over each FASST region. Bar plots: totals for selected world regions and global total. Error bars represent the 95% CI on the RR from the exposure-response function by Burnett et al. (2014)



Premature mortalities from O₃

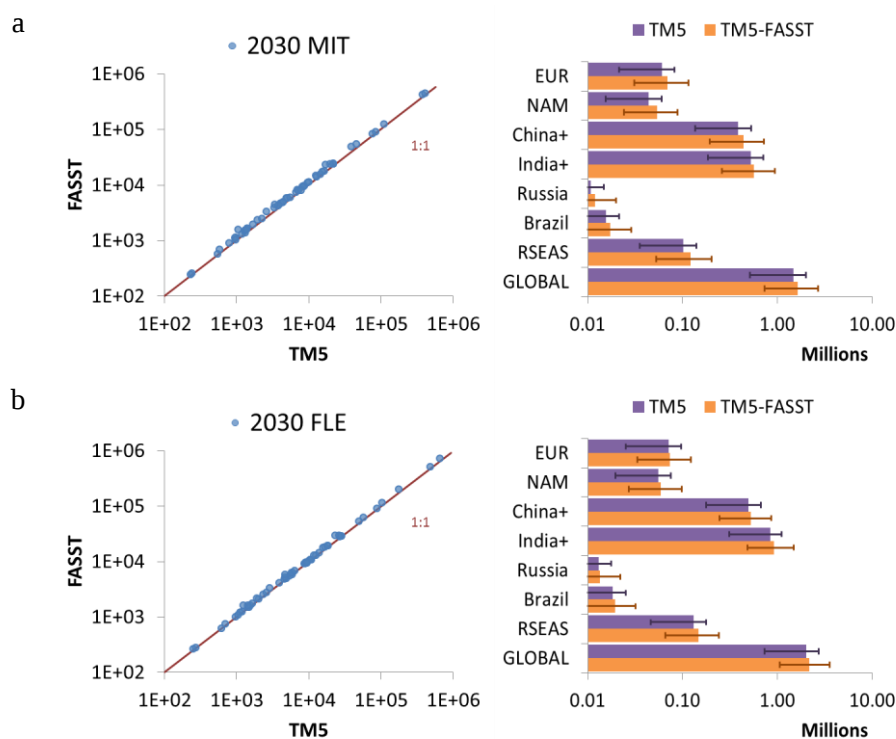


Figure 11: FASST versus TM5 premature mortalities from exposure to O₃ for MIT(a) and FLE-2030 (b) scenarios (see text). Dots: aggregated over each FASST region. Bar plots: totals for selected world regions and global total. Error bars represent the 95% CI on the exposure-response function (Jerrett et al., 2009)

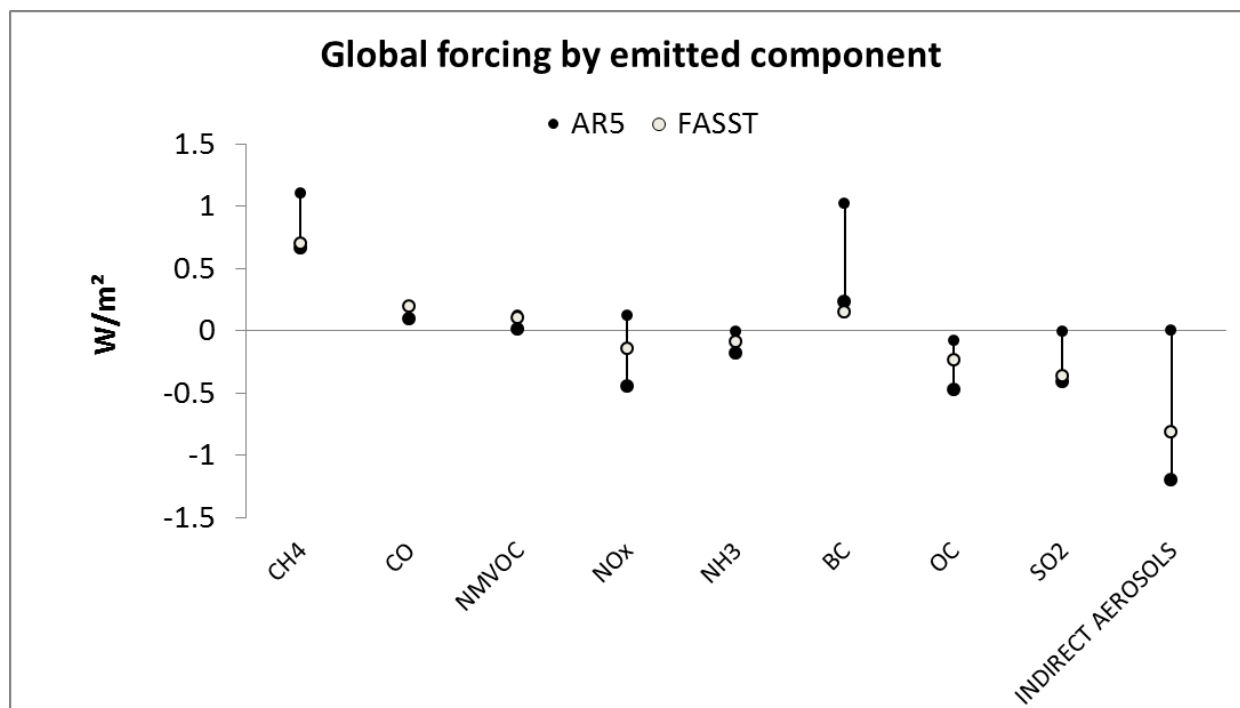


Figure 12: Global anthropogenic radiative forcing by emitted component, from TM5-FASST forcing efficiencies applied on RCP (year 2000 anthropogenic emissions), and range of best anthropogenic forcings from AR5 (change over period 1750 – 2011)

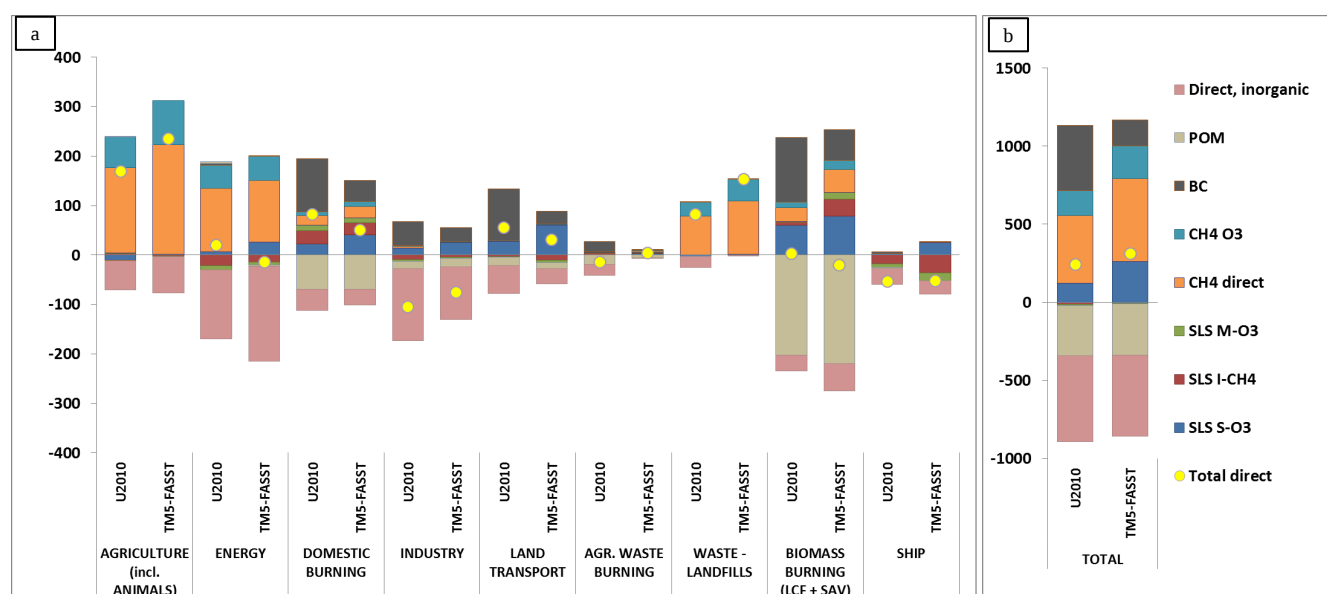


Figure 13: Year 2000 radiative forcing from Unger et al. (2010), based on EDGAR year 2000 emissions and from TM5-FASST applied to RCP year 2000 (a) break-down by sector and by forcing component; (b) total over all sectors.

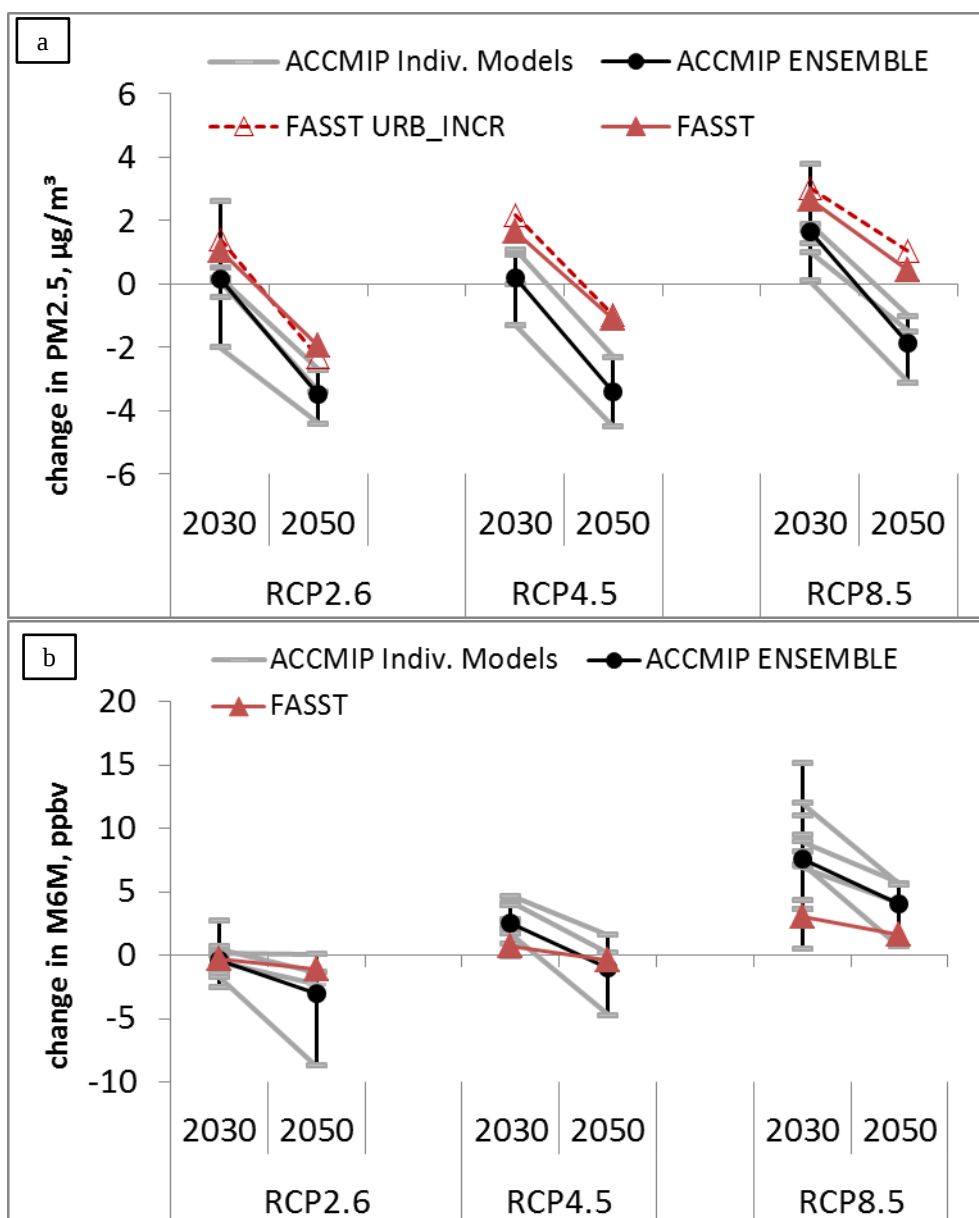


Figure 14: Global population-weighted differences (scenario year minus year 2000) (a) in annual mean PM_{2.5} concentrations and (b) in O₃ exposure metric M6M for 3 RCP scenarios in each future year, from the ACCMIP model ensemble (Silva et al., 2016) (black symbols and lines) and TM5-FASST_v0 (red symbols and lines). FASST URB_INCR: including the urban increment correction. Grey symbols: results from individual ACCMIP models. Grey lines connect results from a single model. Not all models have provided data for all scenarios. ACCMIP error bars represent the range (min, max) across the ACCMIP ensemble.

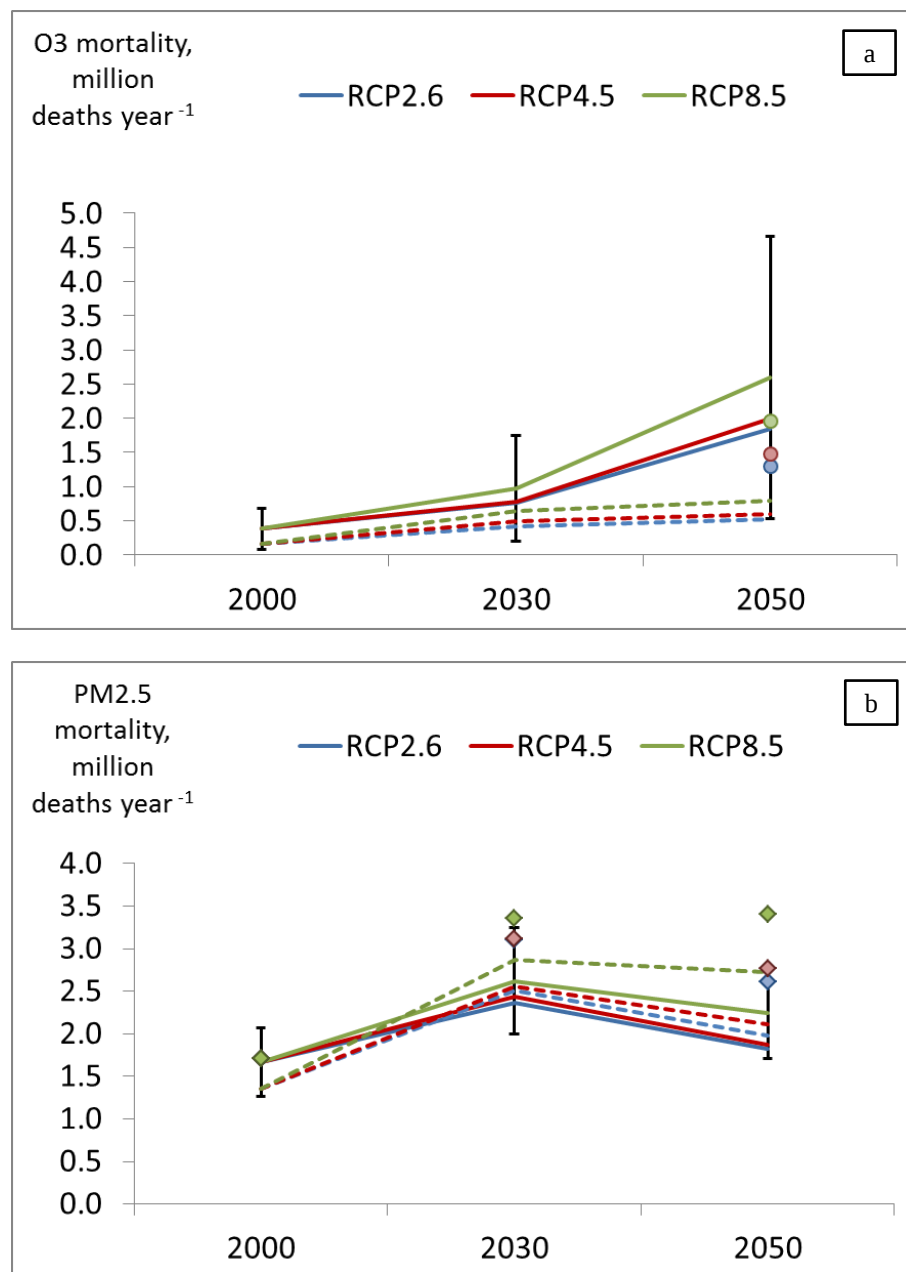


Figure 15. Trends in global burden on mortality of ozone (a) and PM_{2.5} (b) from year 2000 to 2050 from the ACCMIP multi-model ensemble (Silva et al., 2016) (full lines) and TM5-FASST (dashed lines) for 3 RCP scenarios. The error bar on the year 2000 is the ACCMIP 95% CI including uncertainty in RR and across models. CI for 2030 and 2050 were not provided by ACCMIP, we use here the same relative error as for year 2000. Dots (O₃ mortality): adjusted TM5-FASST ozone mortalities for RCP 2050, using baseline respiratory mortalities consistent with Silva et al. (2016). Diamonds (PM_{2.5} mortality): TM5-FASST estimate including the urban increment parameterization